ESTIMATION OF AEROSOL CONCENTRATION IN A NUCLEAR REACTOR CONTAINMENT AFTER AN ACCIDENT*

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ABSTRACT

Results are presented which demonstrate that the particle size distribution of the form:

$$dn/dV = AV^{-2}$$

(where n is number concentration, V is particle volume, and A is related to the maximum and minimum particle volumes in the distribution, according to:

$$A = 1/(V_{min}^{-1} - V_{max}^{-1});)$$

does describe aerosols of interest in nuclear safety. Information is also presented regarding the dependence of V_{min} on environment; aerosol age seems to be the most important variable. The effect of the increase in V_{min} with time on the concentration-time behavior in a containment was estimated. Steam condensation experiments demonstrated that particle deposition occurs by thermophoresis in condensing steam. Particle growth and enhanced settling also must occur in condensing steam. The effect of deposition by thermophoresis on the concentration-time behavior in a containment was estimated. Finally, some estimates of aerosol particle concentration vs time in water-cooled reactor containments resulting from all the growth and deposition processes considered, are presented.

An ultimate purpose of nuclear safety research is to estimate possible radiological doses to citizens. The estimation of the dose from radioactive smoke requires a means of estimating the mass concentration of aerosol in a containment vessel vs time after the accident.

It is anticipated that smoke particles from a water-cooled reactor accident would be similar to those shown in Fig. 1 which is an electron-micrograph of particles of oxides of the constituents of stainless steel formed in a steep temperature gradient in the vicinity of an electric spark between tungsten and stainless steel in dry air. The particles are small (generally 0.01-1 micron), agglomerated, of various sizes and complicated shapes.

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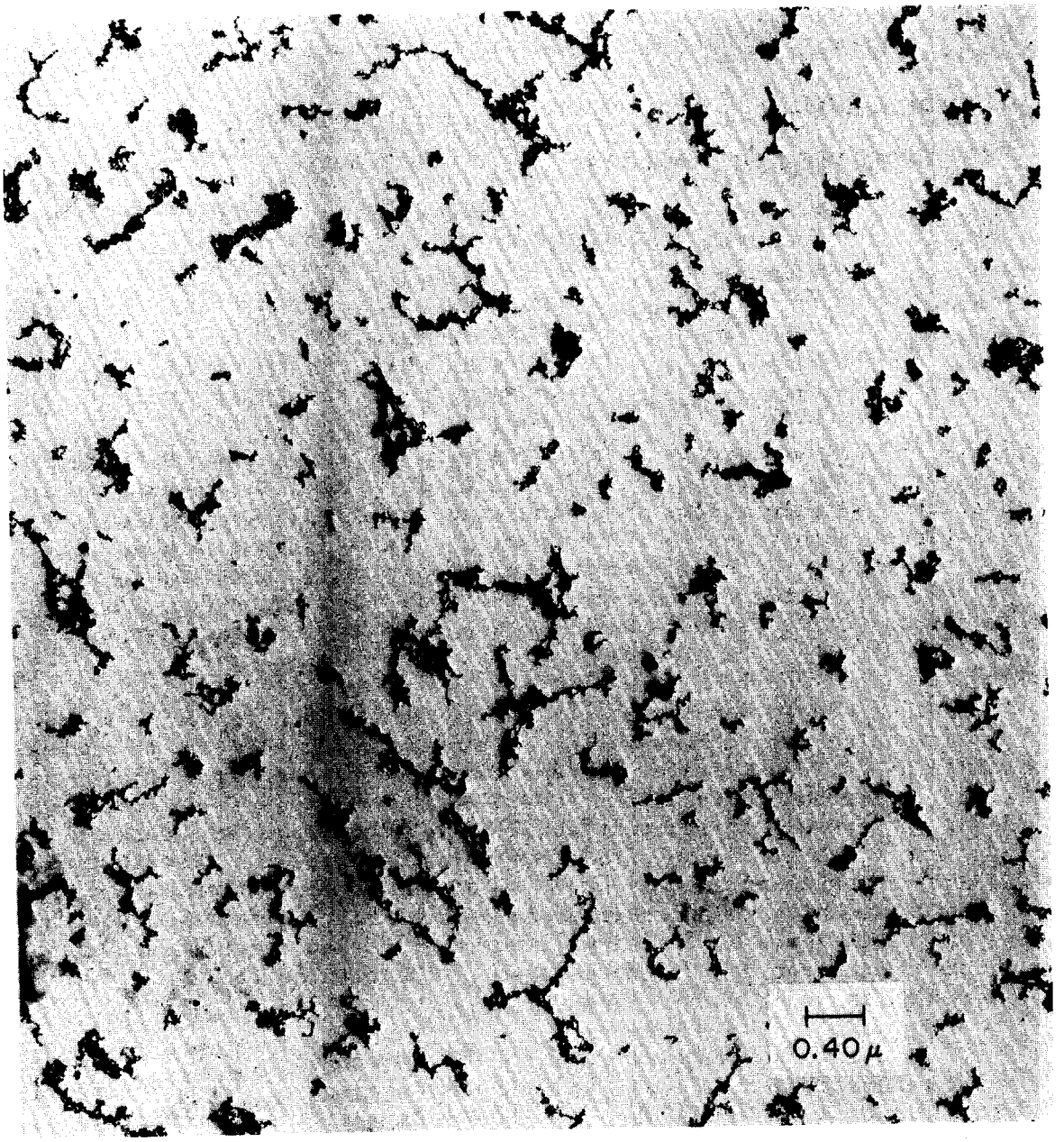

Figure 1

Dry Stainless Steel Oxide Aerosol

Our earlier, reported efforts[1-3] have demonstrated a simple means of estimating the mass concentration of such an aerosol as a function of time and as a consequence of agglomeration and settling. An example of this fit to data is shown in Fig. 2. This calculation was based on an assumed steady-state size distribution function. In terms of particle volume, V, this number concentration, n, distribution function is:

$$[1/n(t)][\partial n(v,t)/\partial V] = AV^{-2} \tag{1}$$

The corresponding volume fraction, ϕ, distribution is

$$[1/\phi(t)][\partial \phi(v,t)/\partial V] = BV^{-1} \tag{2}$$

where

$$A = 1/(V_{min}^{-1} - V_{max}^{-1}) \tag{3}$$

and

$$B = 1/\ln [V_{max}/V_{min}]. \tag{4}$$

V_{max} and V_{min} are, respectively, the maximum and minimum particle volumes in the distribution. The particle volume, V, was defined so that the particle volume times the accepted density of the particle substance would be equal to the particle mass. It was predicted in the reported studies[2,3] that V_{min} was important in determining the concentration-time behavior resulting from coagulation and settling (as shown in Fig. 3) but that V_{max} was not very important (as shown in Fig. 4).

We wished to demonstrate that this[3] simple and versatile means of estimating mass concentration vs time is quite adequate to nuclear safety needs. We therefore became committed to the tasks of: (1) illustrating conclusively that the distribution functions above do describe aerosols of interest and(2) determining the dependence of V_{max} and V_{min} (i.e., the parameters which describe the distribution) on environmental parameters such as initial concentration, age and Reynolds number. We also wished to determine the effects of condensing steam (which could be prevalent in a water-cooled reactor accident) on the concentration vs time behavior. These three tasks will be discussed in order.

The first experimental setup is diagrammed in Fig. 5. Electric spark generated aerosol was produced near one end of a tube; clean air was blown past the spark down the tube. In the first experiment, the tube was 4 inches in diameter, the air flow rate was 1 cu ft per min and the sample of aerosol was taken five feet down the tube. The aerosol sample was diluted and then analyzed with a system of particle sizing instruments. The total number concentration was measured with a condensation nuclei counter,[4] the total mass concentration was measured

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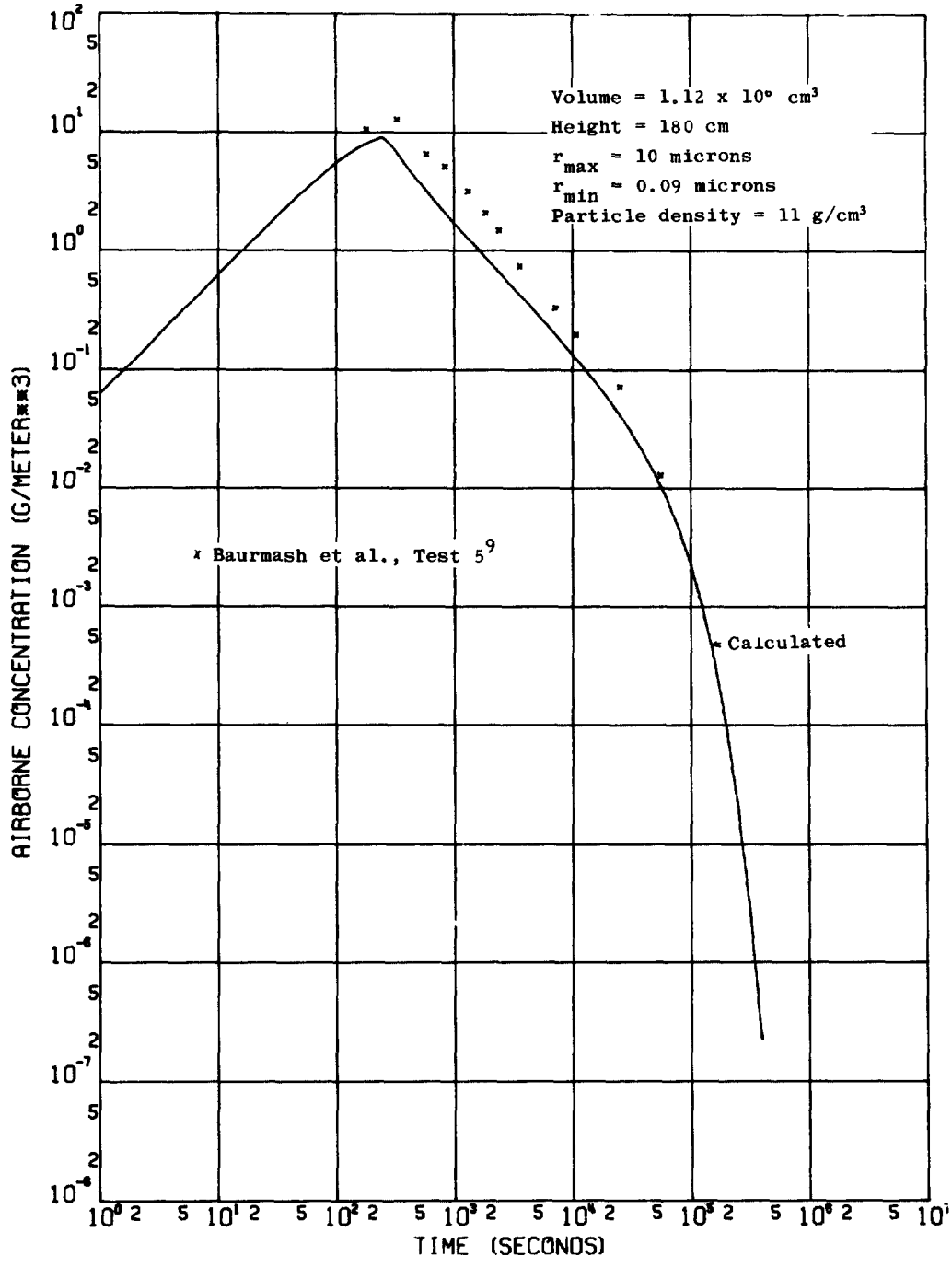

CALCULATED AND OBSERVED CONCENTRATION VS TIME FOR A UO_2 AEROSOL

Figure 2

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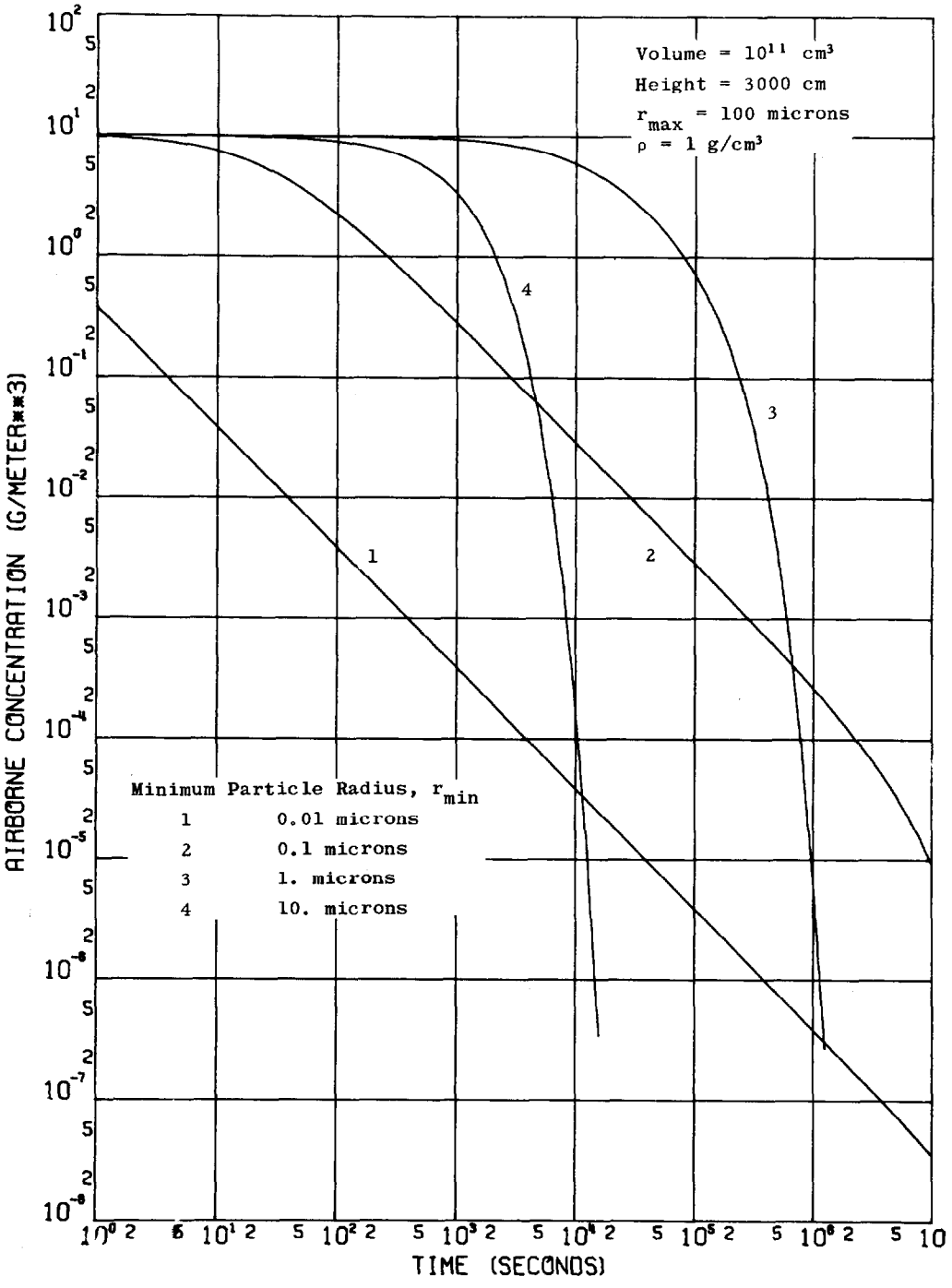
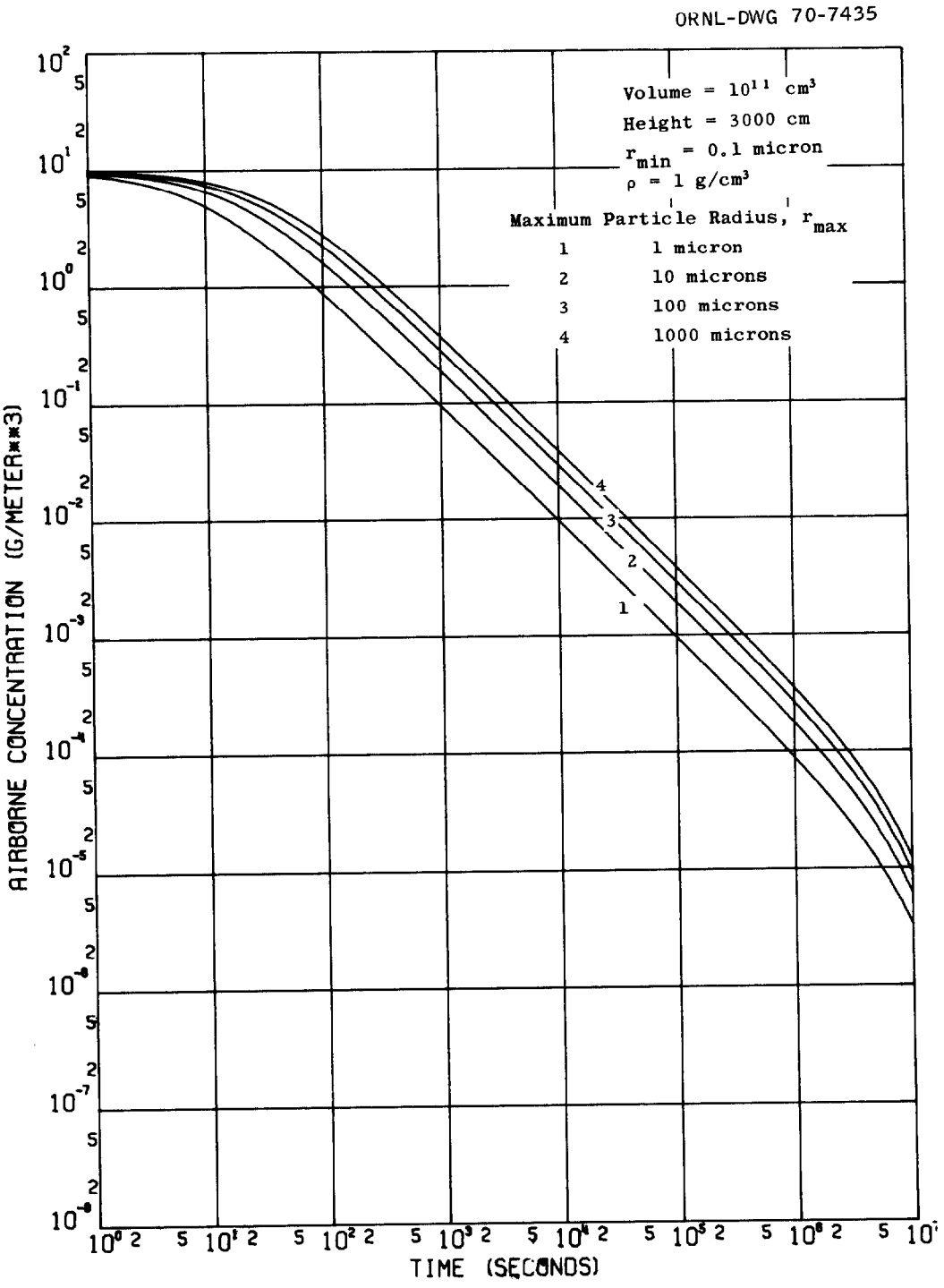


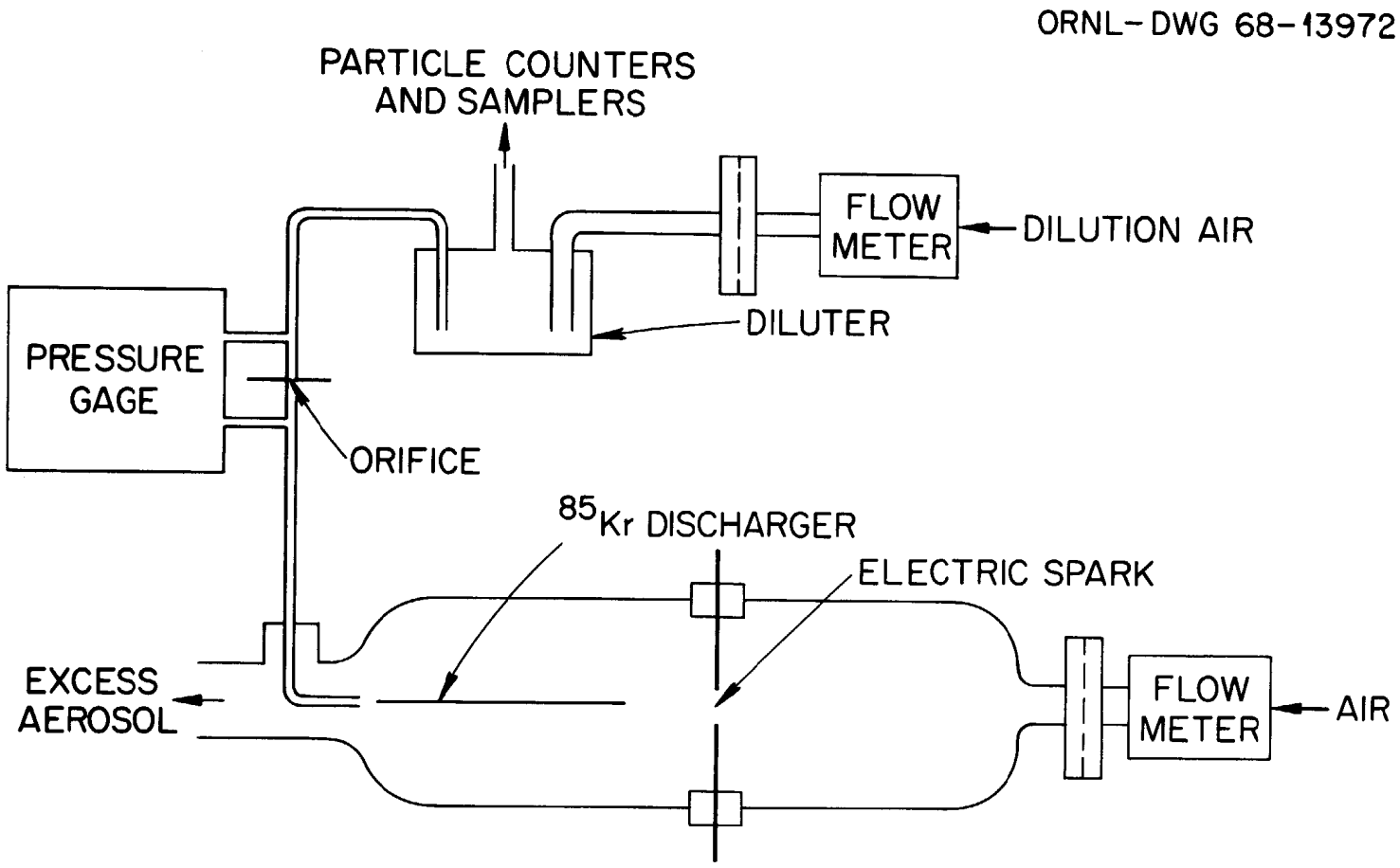
EFFECT OF MINIMUM PARTICLE SIZE

Figure 3



EFFECT OF MAXIMUM PARTICLE SIZE

Figure 4



Apparatus to Measure Particle Size Distribution of Young Spark-Generated Aerosol.

Figure 5

as the total radioactivity (^{51}Cr) collected on all the plates and back-up filter of a low-pressure impactor.[5] The size distribution was measured with four instruments: an electrostatic particle counter[6] (range about 0.015 to 1 μm diameter), an optical particle counter[7] (range about 0.03 to 10 μm diameter), and a low pressure impactor[5] (range about 0.01 to 2 μm diameter); sizing and counting was also done on electronmicrographs of particles precipitated with a thermal precipitator.[8]

Some of the data interpretation needs to be described. The low pressure impactor provides a value of the total mass concentration and values of the fraction of the total mass concentration associated with several particle diameter ranges. These mass concentration values were converted to volume fraction values by dividing by 5.35 g/cm^3 (the weighted average density for the mixture of oxides corresponding to stainless steel). This procedure defines these experimental values of volume such that volume times the actual particle substance density will be equal to mass. The particle sizing on the electron micrographs provided us with a distribution of agglomerate lengths (i.e., the longest straight line across each agglomerate particle). To achieve an estimate of particle volume (again defined so that volume times density equals mass), we considered a simplified equivalent shape for these agglomerates; a cylinder 0.05 μm in diameter and of the measured length. This enabled us to estimate the volume of each particle and to derive a distribution in terms of particle volume. Normalizing factors were determined for the electrostatic and optical counter data to allow those data to conform. The particle diameter values from the optical counter were all multiplied by 3.0, from the electrostatic particle counter by 0.5. The optical counter normalization agrees well with that found by others[9] for dark colored, opaque particles. All other manipulations with the data were according to Eqs. (1-4) or equations derived[3] therefrom.

The results of this data interpretation are shown in Figs. 6 and 7. Figure 6 is a number concentration distribution, dn/dV, vs particle volume V. Figure 7 is the corresponding volume fraction distribution, $d\phi/dV$ vs V. The values for the total number concentration and total volume fraction concentration, which are noted on Figs. 6 and 7, complete the aerosol description.

The dashed lines on Fig. 6 and 7 represent the best fit of the theoretical distributions (Eqs. 1-4) to the data. This fit was obtained as follows. From Eqs. (2) and (4):

$$\ln (V_{max}/V_{min}) = \phi/(d\phi/dV)_{V = 1} \qquad (5)$$

where $(d\phi/dV)_{V = 1}$ signifies the value of the volume fraction distribution function at V equals unity. ϕ and $(d\phi/dV)_{V = 1}$ can be taken directly from the data, hence the ratio V_{max}/V_{min}) can be calculated. Next, from Eqs. (1) and (3):

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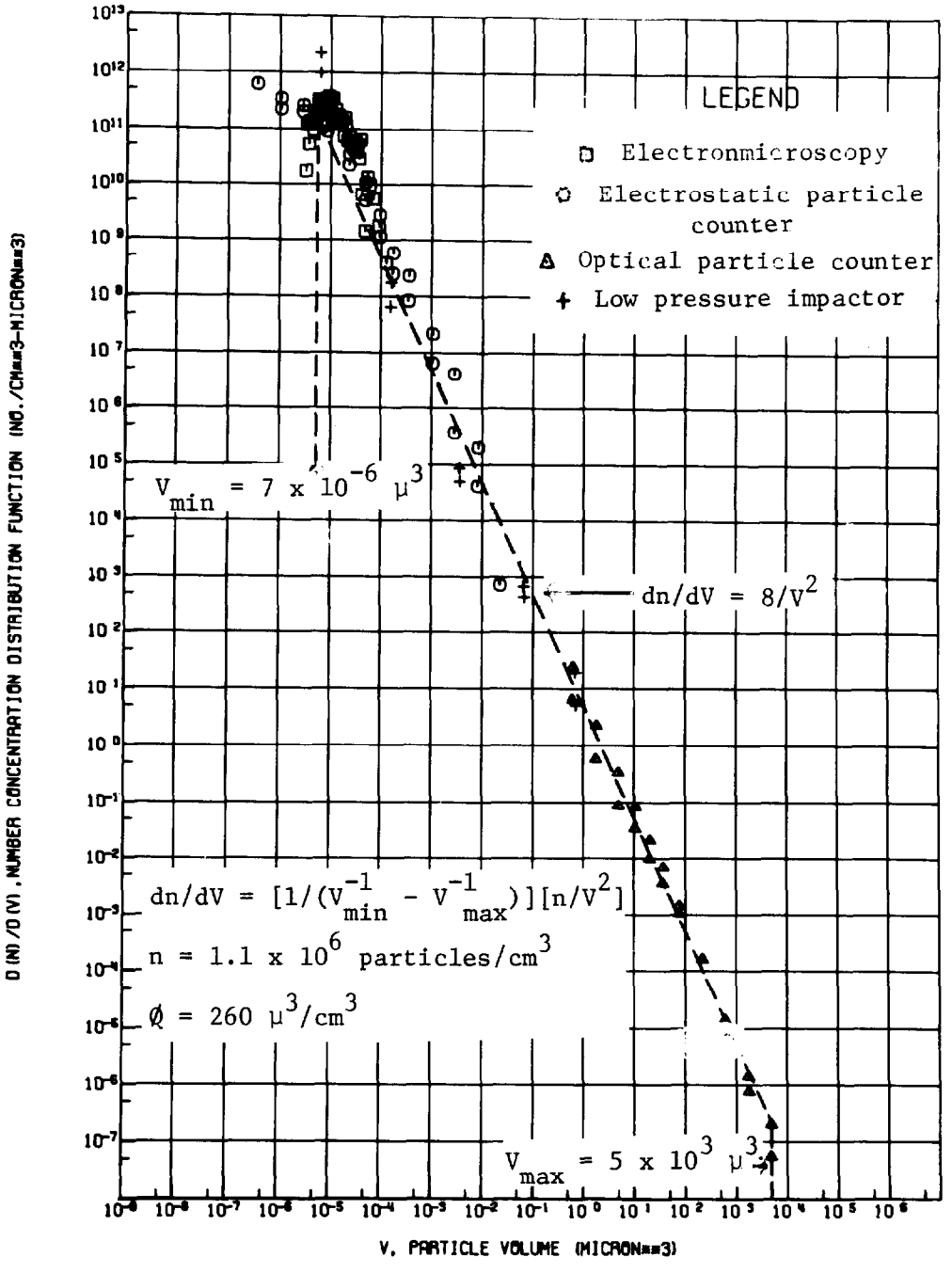

A NUMBER CONCENTRATION DISTRIBUTION OF A STAINLESS STEEL OXIDE AEROSOL

Figure 6

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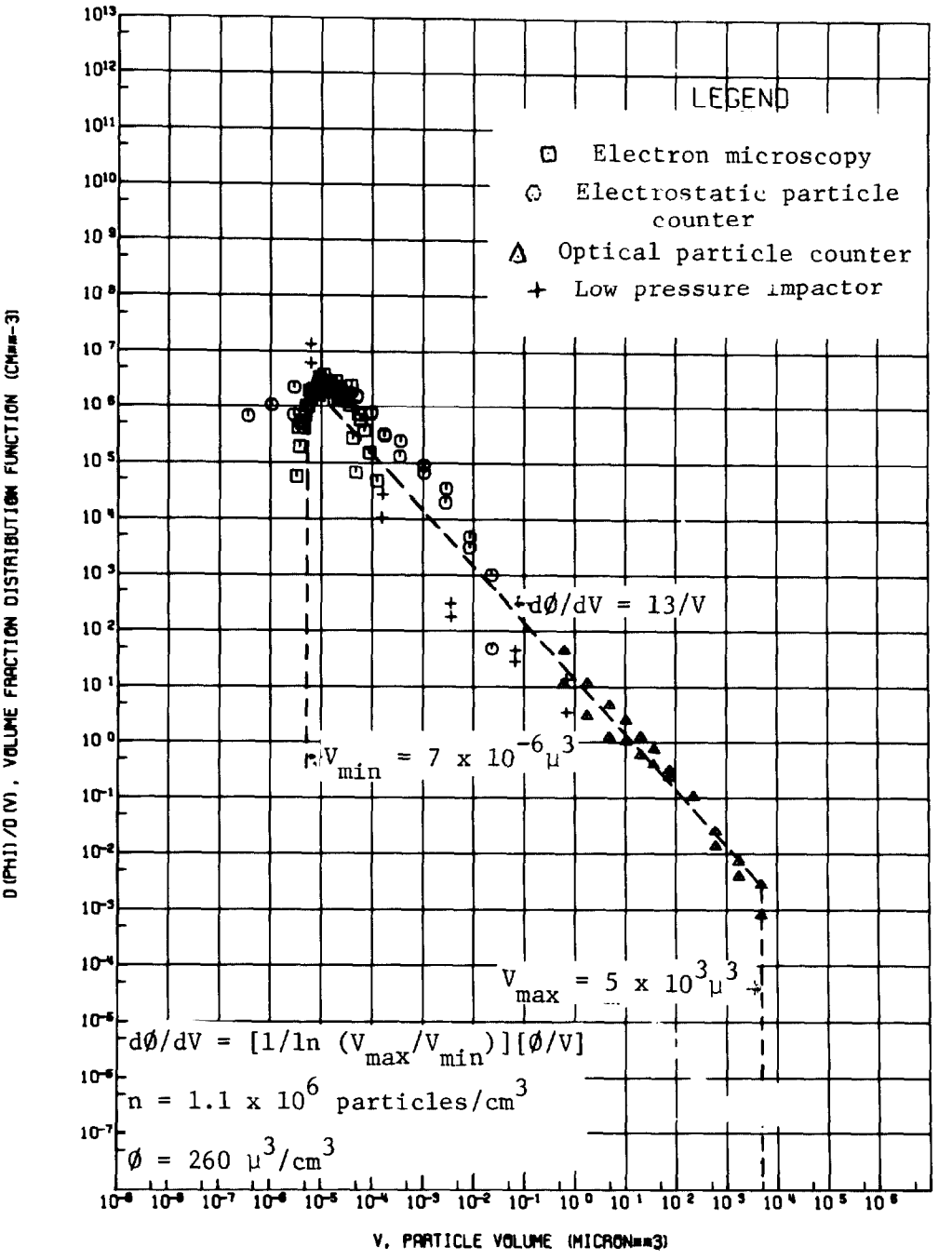


A VOLUME FRACTION DISTRIBUTION OF A STAINLESS STEEL OXIDE AEROSOL

Figure 7

$$V_{min}^{-1} = V_{max}^{-1} + n/(dn/dV)_{V=1} \approx n/(dn/dV)_{V=1} \quad (6)$$

where $(dn/dV)_{V=1}$ is the number concentration distribution at V equals unity. The indicated simplification is possible since V_{max}^{-1} is clearly (from the data) much smaller than V_{min}^{-1}. So with Eq. (6) V_{min} can be calculated. V_{max} can now be obtained by putting the value of V_{min} into Eq. (5). The values of V_{max}, V_{min}, $(dn/dV)_{V=1}$ and $(d\emptyset/dV)_{V=1}$ are all that is needed to draw the dashed lines on Figs. 6 and 7; hence, the theoretical equation is fitted to the data on the basis of adjustment of V_{max} and V_{min}.

Figures 6 and 7 do in fact illustrate that the distribution functions (Eqs. 1-4) do describe aerosols of interest; the first task has therefore been accomplished.

The second task was to determine the dependence of V_{max} and V_{min} on environmental parameters, especially initial concentration, age and Reynolds number. The experimental setup shown in Fig. 5 was employed By varying the tube length, tube diameter and air flow one can achieve a systematic variation in aerosol age, Reynold's number and initial concentration. In our tests the tube diameter was about 4 inches; we have varied the tube length and the air flow rate (but not the diameter); the tube lengths from 1 ft to 5 ft to 50 ft and the air flow rate between 0.15 and 14 cu ft per min. This gave us Reynold's numbers from less than 100 to 5600, aerosol ages from less than a second to 30 minutes or so and concentrations of 5×10^{-5} to 5×10^{-3} g/m^3. We also varied the humidity; in most tests the air was dry, in some it was near 100 relative humidity. For each test a size distribution was determined (as in Figs. 6 and 7), and the values of V_{max} and V_{min} were determined as described above.

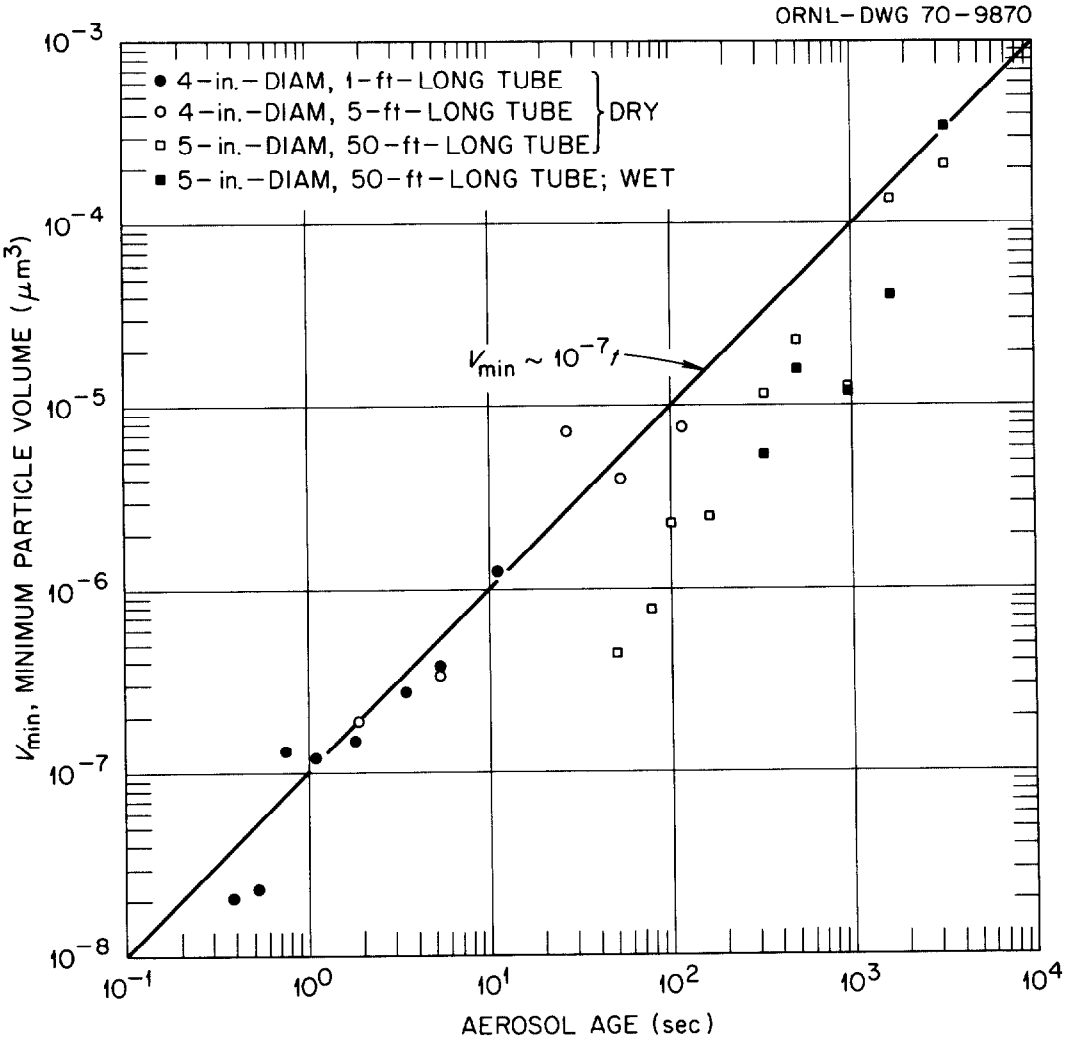
Figure 8 is a plot of all the V_{min} values vs aerosol age. All the V_{min} values appear to correlate with this single parameter according to

$$V_{min} = 10^{-7}\ t, \text{ for t greater than 0.4 seconds} \quad (7)$$

where V_{min} is in μm^3 and t (time) is in seconds. This suggests that humidity, initial concentration and Reynold's number are relatively unimportant parameters.

Figure 9 is a series of number distributions for different aged aerosols; it demonstrates, with the actual data, that a major effect of age is indeed a continual increase in V_{max}. Figure 10 shows the volume fraction distribution of the same data shown in Fig. 9; it also shows the increase in V_{min} with age.

The V_{max} values (in all the experiments) varied between 200 and 104 μm; they tended to decrease with time. Since V_{max} does not significantly change the concentration-time behavior, its dependence on time was not pursued.



Minimum Particle Volume as a Function of Aerosol Age.

Figure 8

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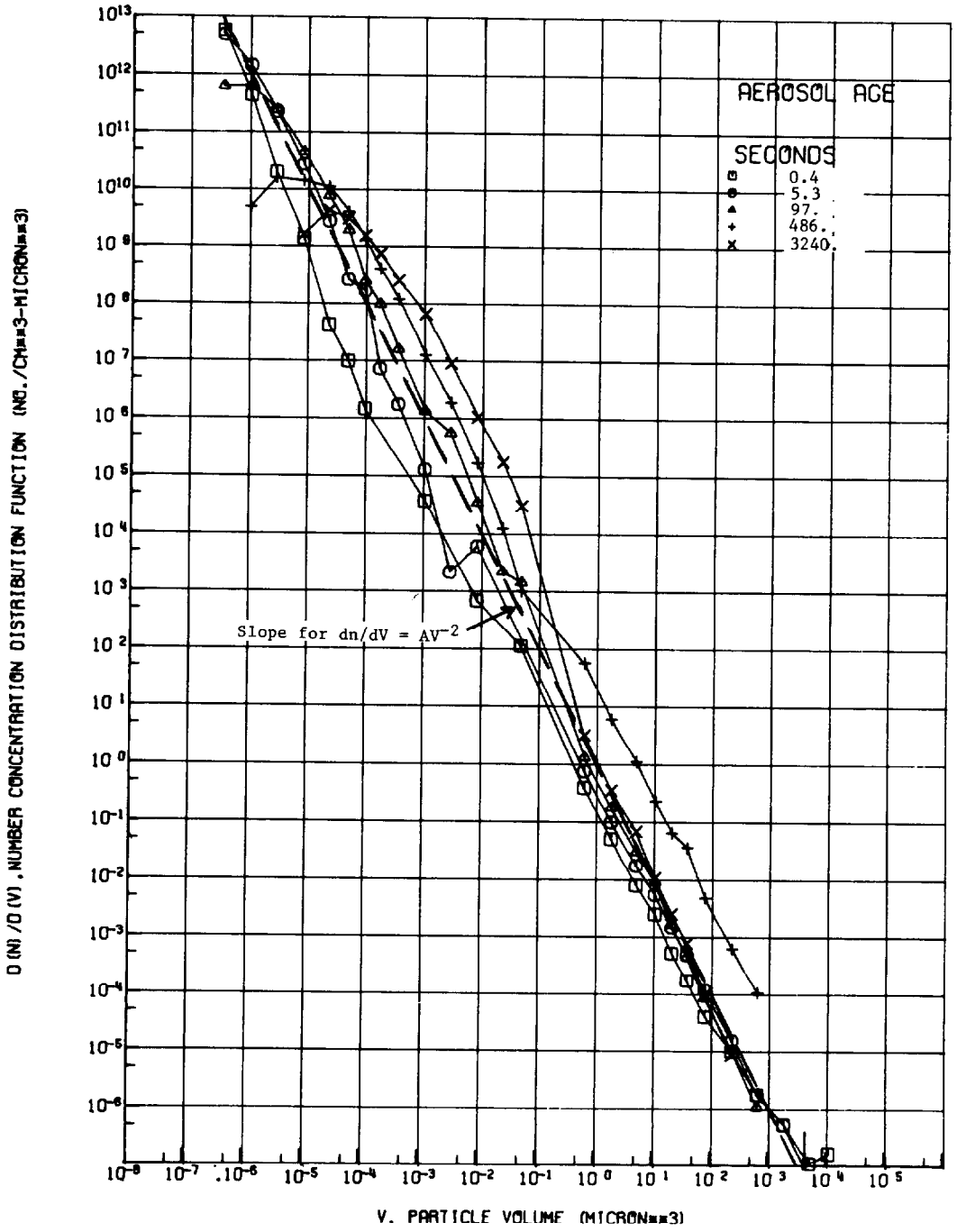
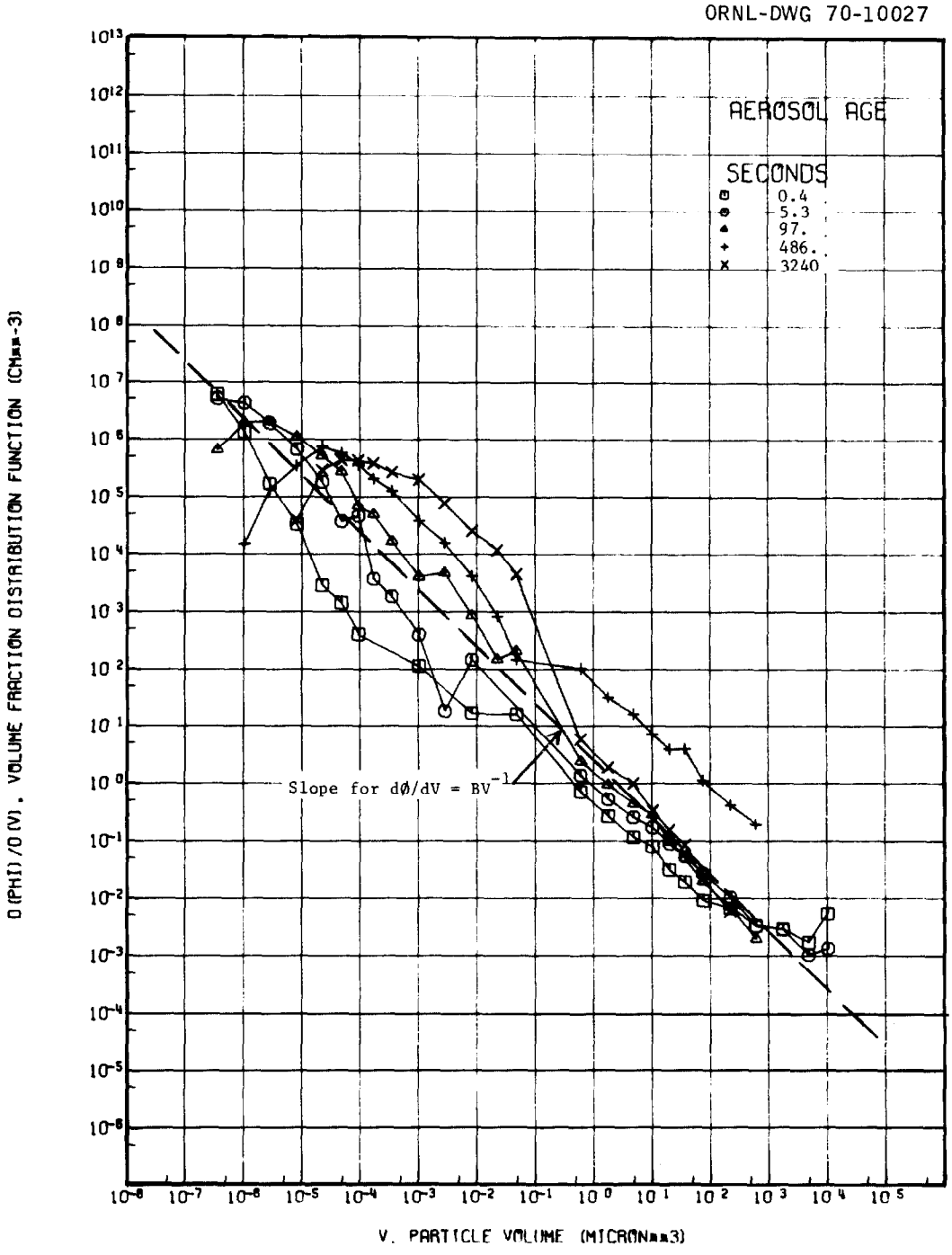


EFFECT OF AGE ON NUMBER CONCENTRATION DISTRIBUTION

Figure 9



EFFECT OF AGE ON VOLUME FRACTION DISTRIBUTION

Figure 10

It is of interest to determine how importantly the V_{min} vs time dependence affects the concentration vs time behavior resulting from agglomeration and settling. To suggest the importance, the SMOKLEAR code[3] was modified to include an incrementally changing V_{min} value according to Eq. (7). A comparison of the calculated concentration-time behavior, with and without the V_{min} time dependence is shown in Fig. 11. As can be seen, the effect is significant.

So the above results at least partially determine the dependence of V_{max} and V_{min} on environmental parameters; the SMOKLEAR calculation of concentration vs time demonstrates the importance of the V_{min} increase with time; the second task is thus partially accomplished.

The third task was to determine the effect of condensing steam on concentration-time behavior. We have pointed out in previously published work[1] that condensing steam should deposit particles and that the rate of deposition should be significant in relation to deposition by settling. The idea was that steam generated by the accident would either condense on cold surfaces or on aerosol particles. If it condenses on cold surfaces, then the flux of water molecules to the cold surfaces will sweep particles by diffusiophoresis. If the steam condenses on particles, the latent heat of condensation will heat the air, the heat will be conducted to cold surfaces and the particles will be swept toward cold surfaces by thermophoresis. The consequences of diffusiophoresis and of thermophoresis are (in a condensing steam system) generally similar.

In a reactor containment after an accident we would expect considerable turbulence; particularly with containment sprays. We would expect therefore that the particle concentration would be uniform, that turbulence would carry particles almost to the walls and that the sweeping action of condensing steam would operate over a short distance, i.e., across a more-or-less stagnant layer.

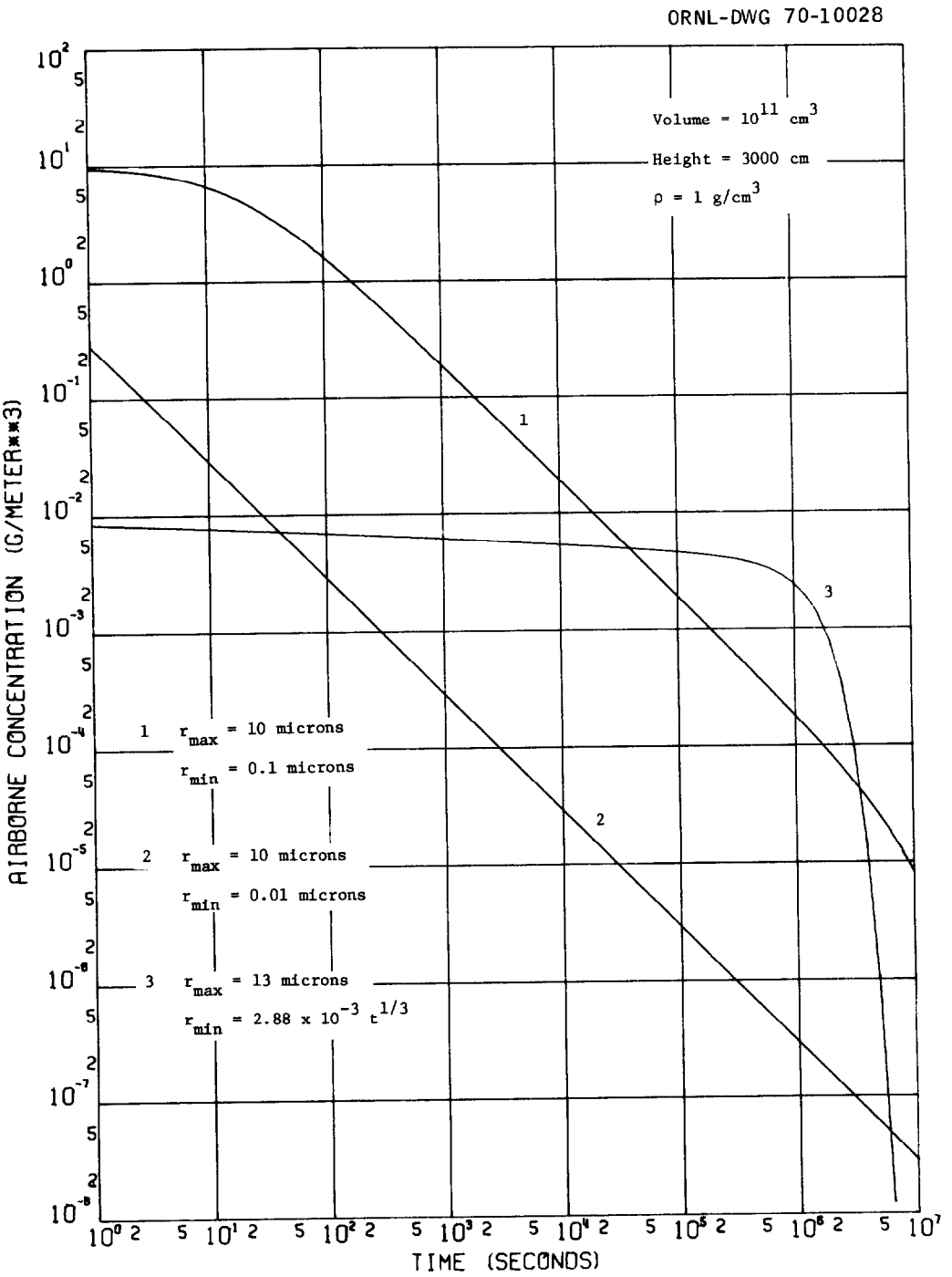
For the case of thermophoresis, the particle deposition velocity across this stagnant layer (at an air temperature near 25°C) is proportional to the temperature gradient;

$$V_T = - 2.6 \times 10^{-4} \, dT/dz \text{ cm/sec.} \qquad (8)$$

The particle size dependence of V_T in the size range of interest is small.[11] By making use of the fact that heat is also conducted at a rate proportional to the temperature gradient,

$$\frac{dq}{dt} = - 2.8 \times 10^{-4} A \, dT/dz, \qquad (9)$$

(where dq/dt is watts, the value 2.8×10^{-4} is the conductivity of air in watts/cm-°C and A is the area for heat transfer (cm^2)); we can eliminate the thermal gradient and (approximately eliminate) the uncertainties associated with the boundary layer thickness, hence:



EFFECT OF INCREASE IN MINIMUM SIZE WITH TIME

Figure 11

$$V_T = \frac{0.929}{A} \frac{dq}{dt} . \quad (10)$$

If the particles in a concentration n particles/cm^3 move toward area A at a velocity of V_T, then the flux of particles to the surface is $V_T n$; in a time increment dt, $V_T nAdt$ particles will be deposited. A similar number of particles, Vdn (where V is the volume of the aerosol) will have been lost from the aerosol phase. The number loss must equal the number deposited, hence:

$$- Vdn = V_T nAdt, \quad (11)$$

therefore

$$\frac{dq}{dt} = n \left(\frac{A}{V}\right) V_T = \left[\frac{0.929}{V}\left(\frac{dq}{dt}\right)\right] n. \quad (12)$$

So the rate of decrease in number concentration should be proportional to the number concentration, proportional to the rate of heat transfer (watts) and inversely proportional to the containment volume.

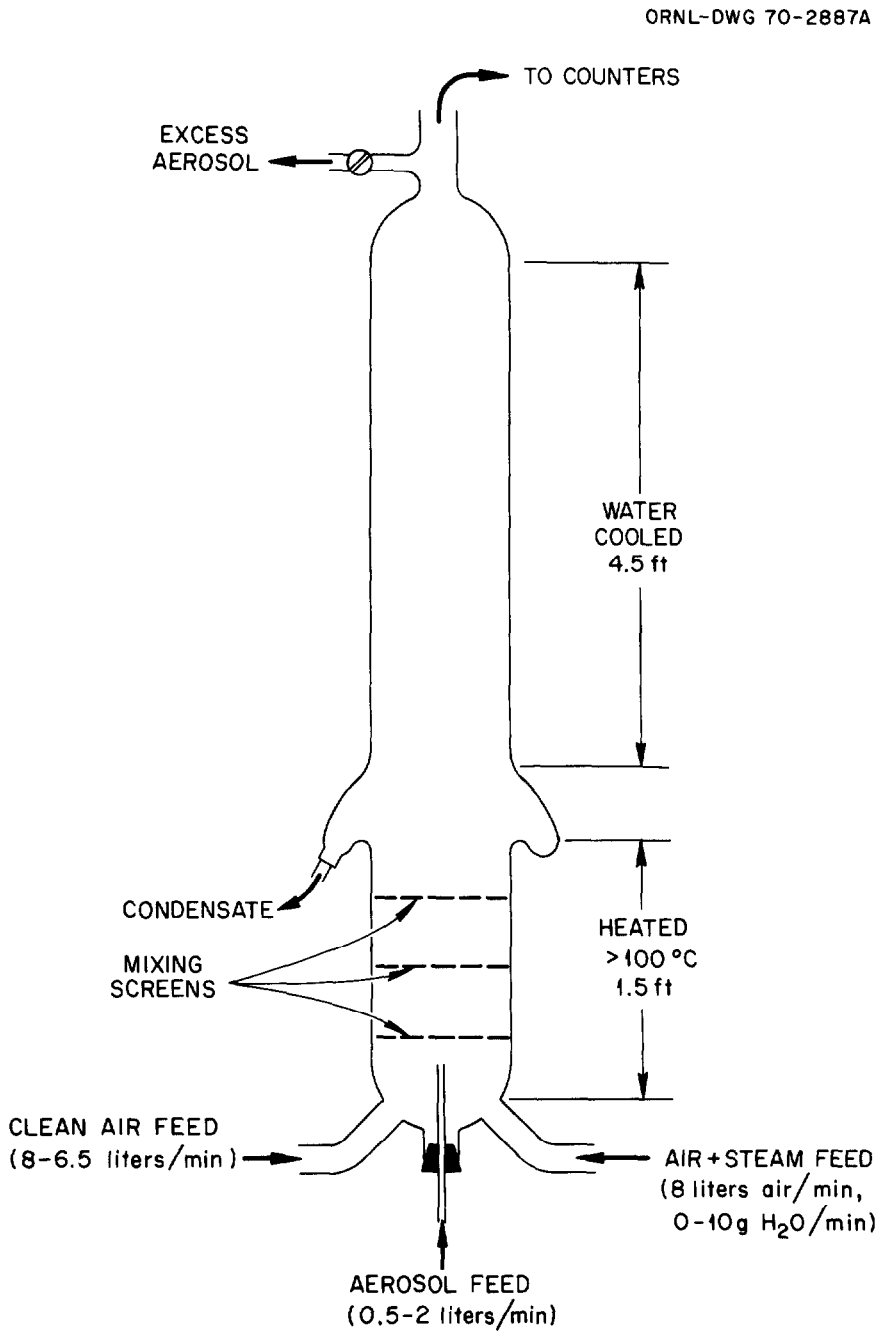
A similar derivation[1,10] for deposition by diffusiophoresis would yield an equation similar to Eq. (12) except the constant would be about half as large; in other words, diffusiophoresis is only half as efficient (in condensing steam) as thermophoresis at sweeping particles.

If the water condenses on the particles (rather than on the walls) one consequence is that thermophoresis will be the sweeping process. Another consequence is that the particles will have grown and being larger will settle faster. So we wished to find out whether steam at low supersaturation would condense mostly on cold walls and therefore sweep particles by diffusiophoresis or mostly on the particles, therefore resulting in the more efficient deposition by thermophoresis as well as enhanced deposition by settling.

The experimental setup is shown in Fig. 12; it is a vertically oriented 4-inch glass pipe; aerosol, air and a steam-air mixture were admitted into the bottom. The lower 18 inches of the pipe contained screens and was heated; the screens mixed the inlet streams and the heat prevented condensation until after mixing. In the top section the aerosol particles were subjected to condensing steam. The effluent aerosols from the top of the condenser were analyzed. The number concentration of the inlet (spark generated stainless steel oxide) aerosols and the steam feed rate (and hence the steam condensation rate) were varied. The upward velocity in all experiments was 3.8 cm/sec.

By an analysis similar to that leading to Eq. (11) it can be shown[10] that if, in this experiment, the water condenses directly on the walls, and diffusiophoresis is the sweeping force, then the ratio of the outlet to inlet concentration, N/N_o, will depend on W, the steam condensation rate (g H_2O/cm^3 aerosol) according to:

$$\log N/N_o = - 385 W. \quad (13)$$



Schematic Diagram of Condenser Scrubber Experiment.

Figure 12

If thermophoresis is the operating process, then

$$\log N/N_o = - 730\ W. \qquad (14)$$

In the case of thermophoresis, the water condenses on particles; in this experiment (with upward aerosol flow at 3.8 cm/sec) the particles may grow enough so that their falling velocity is significant in relation to 3.8 cm/sec. It can be shown[10] that this would lead to an N/N_o dependence on the inlet number concentration as well as the steam condensation rate according to:

$$\log N/N_o = - 730W\ [V_M/(V_M - 4.65 \times 10^5 (W/N_o)^{2/3})]. \qquad (15)$$

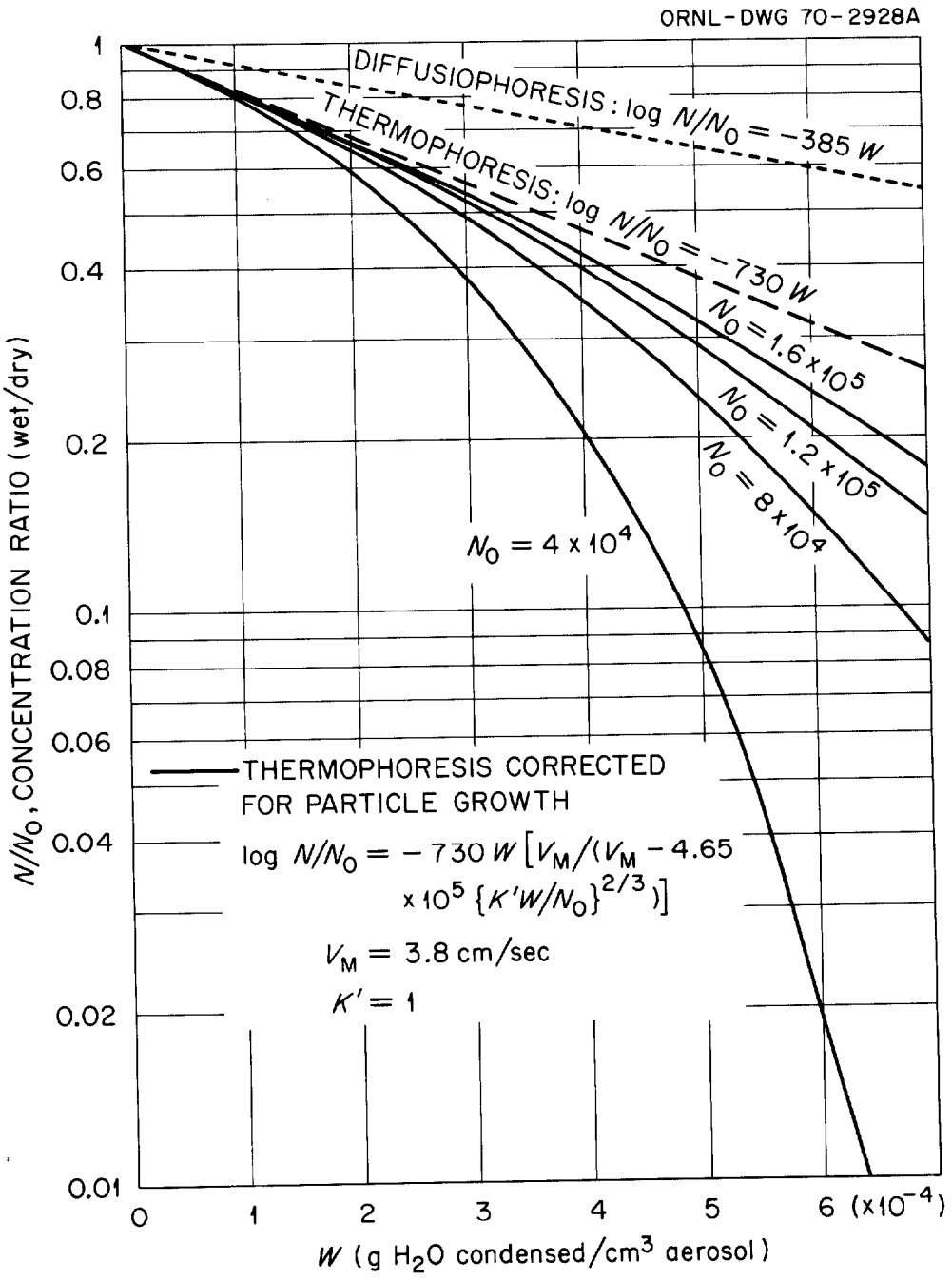
where V_M is the upward velocity of the aerosol medium, 3.8 cm/sec.

These predicted possible behaviors are shown in Fig. 13 as a plot of log N/N_o vs W. Note that thermophoresis is a more efficient process; it removes a larger fraction of the particles with a given steam condensation rate than does diffusiophoresis. In the case of thermophoresis at the lower values of N_o (inlet concentration), where the amount of steam per particle was greater and therefore where more growth of each particle was possible, the additional effect of particle growth to enhance settling (to decrease N/N_o) becomes large.

Figure 14 is a plot of log N/N_o vs W for observed data. The behavior is clearly that identified with thermophoresis and particle growth. At high number concentration, growth is not important, N/N_o follows the thermophoresis line (dashed line on Fig. 14) corresponding to Eq. (14). At lower concentrations the N/N_o values are considerably below the thermophoresis line, as predicted.

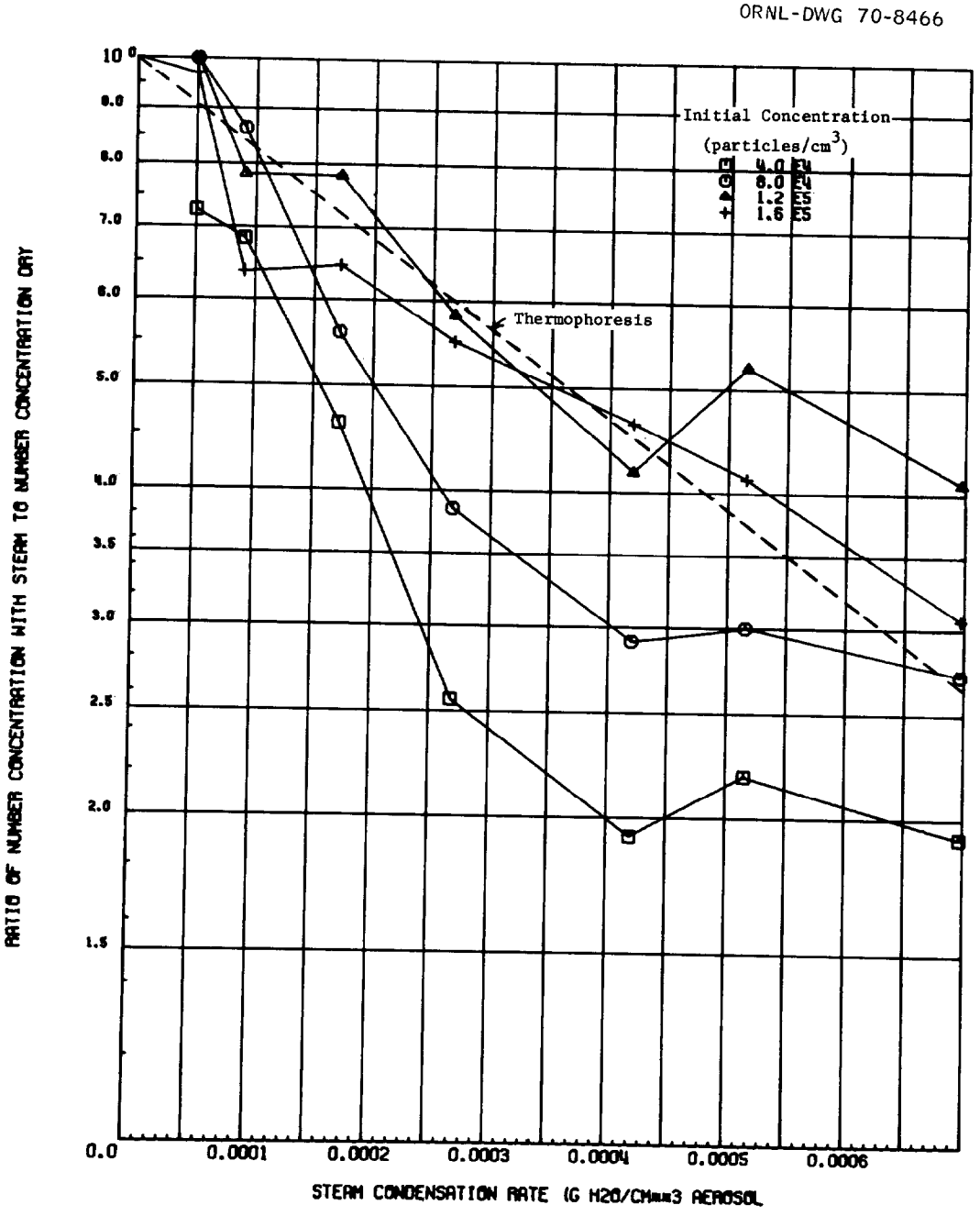
Figures 15 and 16 are, respectively, some number concentration distributions and volume fraction distributions as a function of steam condensation rate. There is a noticeable bulge in the distribution near the large size end of the spectrum due to steam condensation; this also demonstrates that particle growth occurred. These distributions do not necessarily represent what one might get in a large vessel in which steam was condensing: first because these distributions are proably not very accurate; the aerosol particles were, of course, somewhat volatile hence may have changed size somewhat during measurement; second, this experiment was intentionally designed to concentrate the large particles (in order to demonstrate their presence), a large containment vessel would not necessarily function similarly. The results do show the important fact of growth.

We need now to consider what these steam condensation results suggest in regard to an accident aerosol in a water-cooled reactor containment. To do this, we again modified SMOKLEAR[3] to include particle deposition by thermophoresis according to Eq. (12). SMOKLEAR calculates n (number concentration) vs t according to a general differential equation:



Theoretical Increase in Removal Efficiency Due to Growth of Hydrophillic Particles.

Figure 13



OBSERVED PARTICLE REMOVAL IN THE CONDENSER EXPERIMENT

Figure 14

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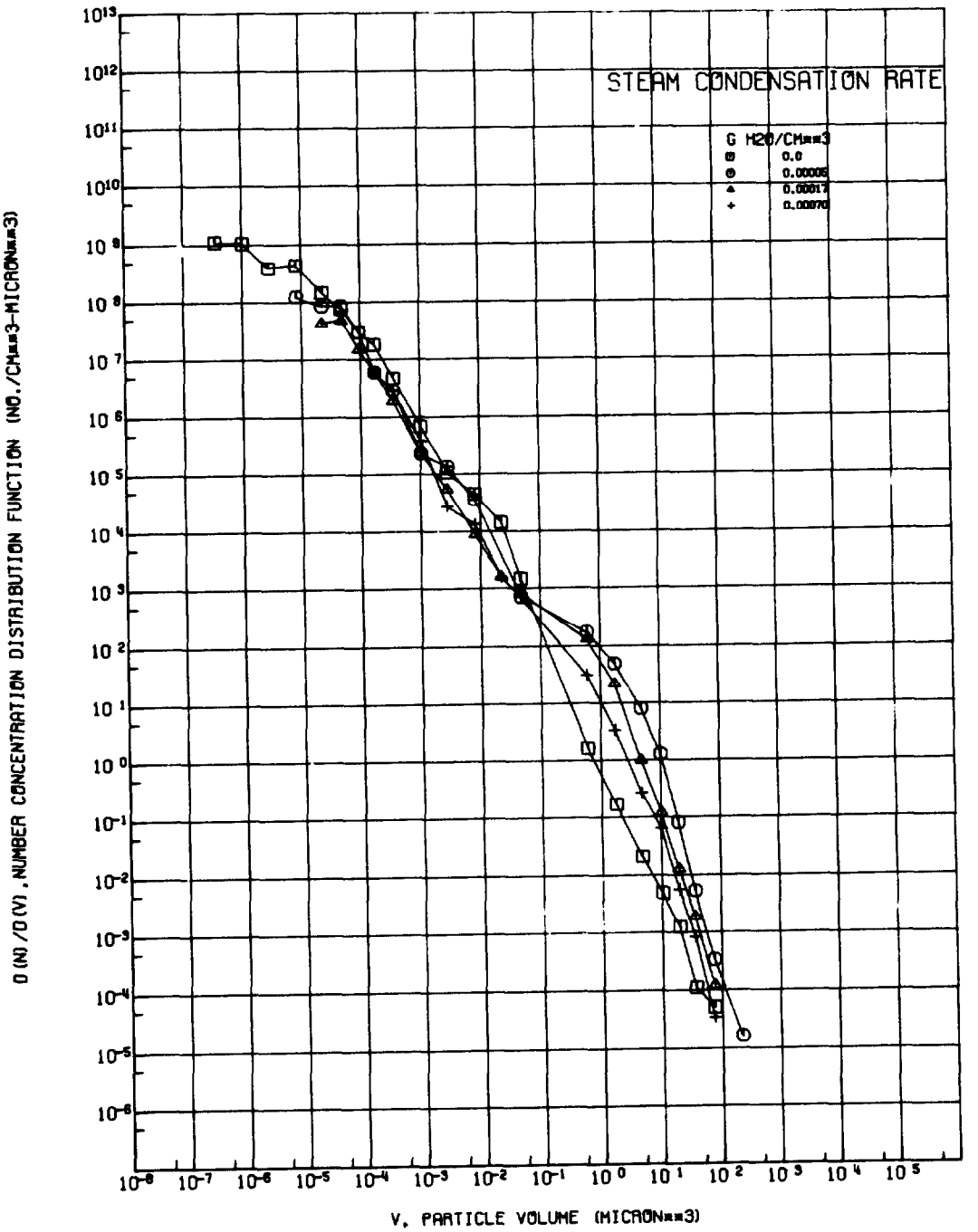
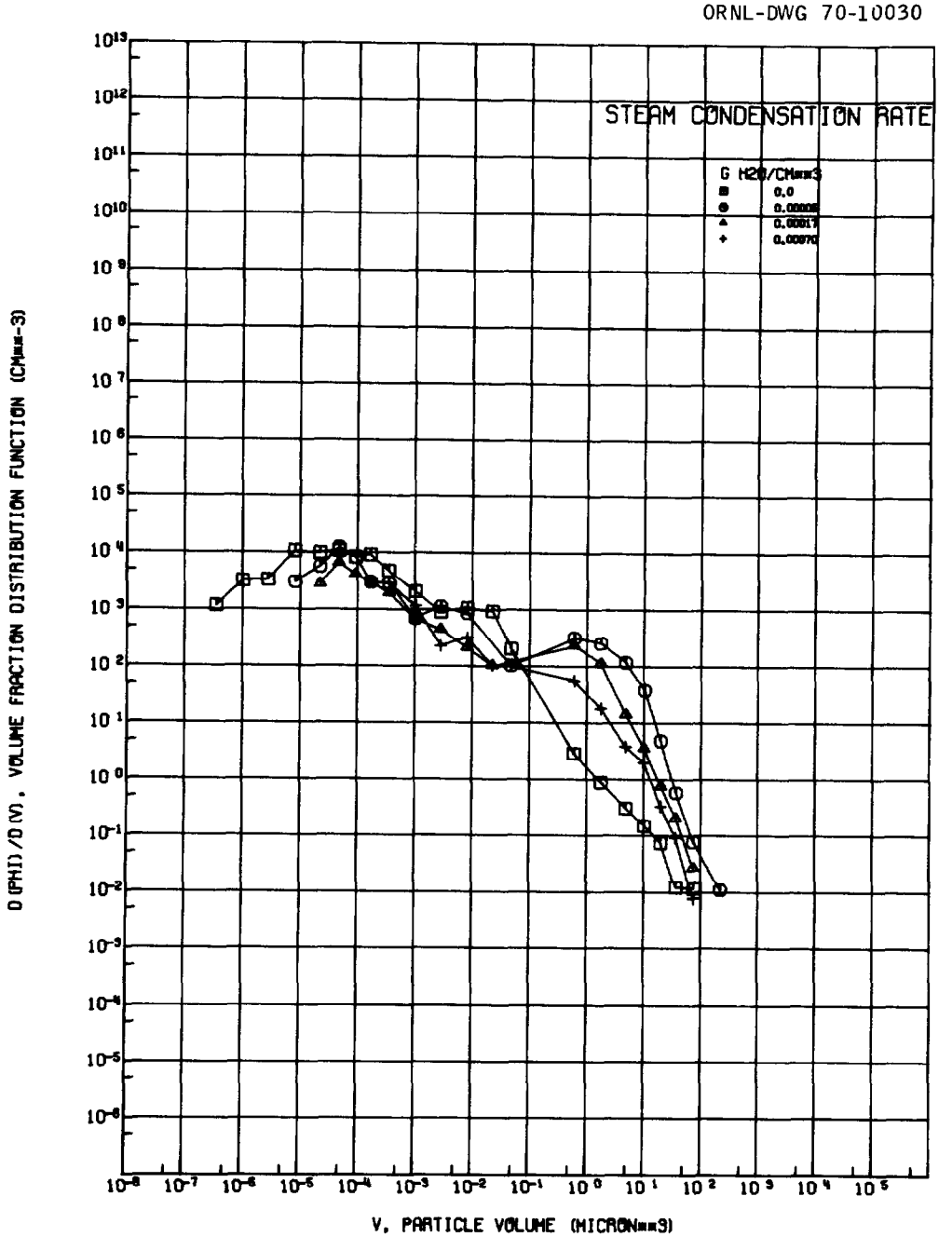


EFFEFT OF CONDENSING STEAM ON NUMBER CONCENTRATION DISTRIBUTION

Figure 15



EFFECT OF CONDENSING STEAM ON VOLUME FRACTION DISTRIBUTION

Figure 16

$$\frac{dn(t)}{dt} = - Kn^2(t) - K^*n(t) + K^{**} \qquad (16)$$

where K is an agglomeration rate constant, K** is a formation term and K*n(t) is a term which is the sum of all the processes considered, the rates of which are proportional to n(t). Hence K* refers to removal by stirred settling, by a recirculating filter and now (after modification) it refers as well to removal by thermophoresis. For dq/dt (in Eq. 12) we used the reactor decay heat; in other words we assumed that the decay heat would produce steam which would later condense in the secondary containment. The decay heat for an illustrative, old core[12] has been calculated to be:

$$q = (585\ t^{0.734} - 3.9t)10^6 \text{ joules} \qquad (17)$$

Hence,

$$\frac{dq}{dt} = (429t^{-0.266} - 3.9)10^6 \text{ watts} \qquad (18)$$

and on substituting into Eq. (11):

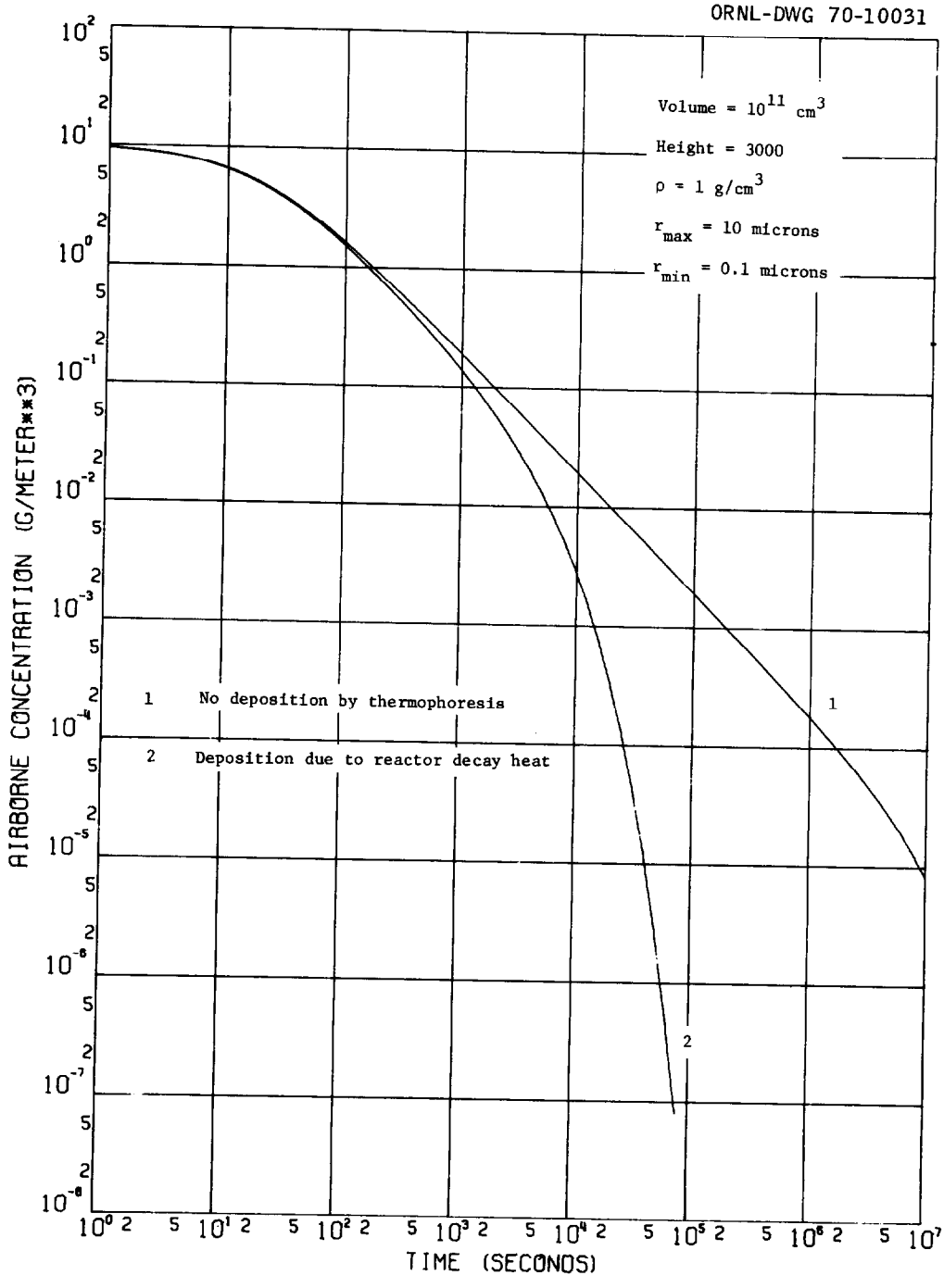
$$\frac{dn}{dt} = n[(\frac{0.929}{V})(429t^{-0.266} - 3.9)10^6]. \qquad (19)$$

The term multiplied by n on the right side of Eq. (18) was added to K* in Eq. (16) and so became part of SMOKLEAR. Of course, this is done incrementally, that is, a new K* is calculated for each time increment.

Figure 17 shows the calculated effect on the concentration-time behavior of deposition by thermophoresis. It is a significant effect and in the helpful direction. There would be, of course, some additional effect of steam condensation on concentration-time behavior, due to enhanced settling resulting from growth. We do not now suggest any quantitative prediction of this effect.

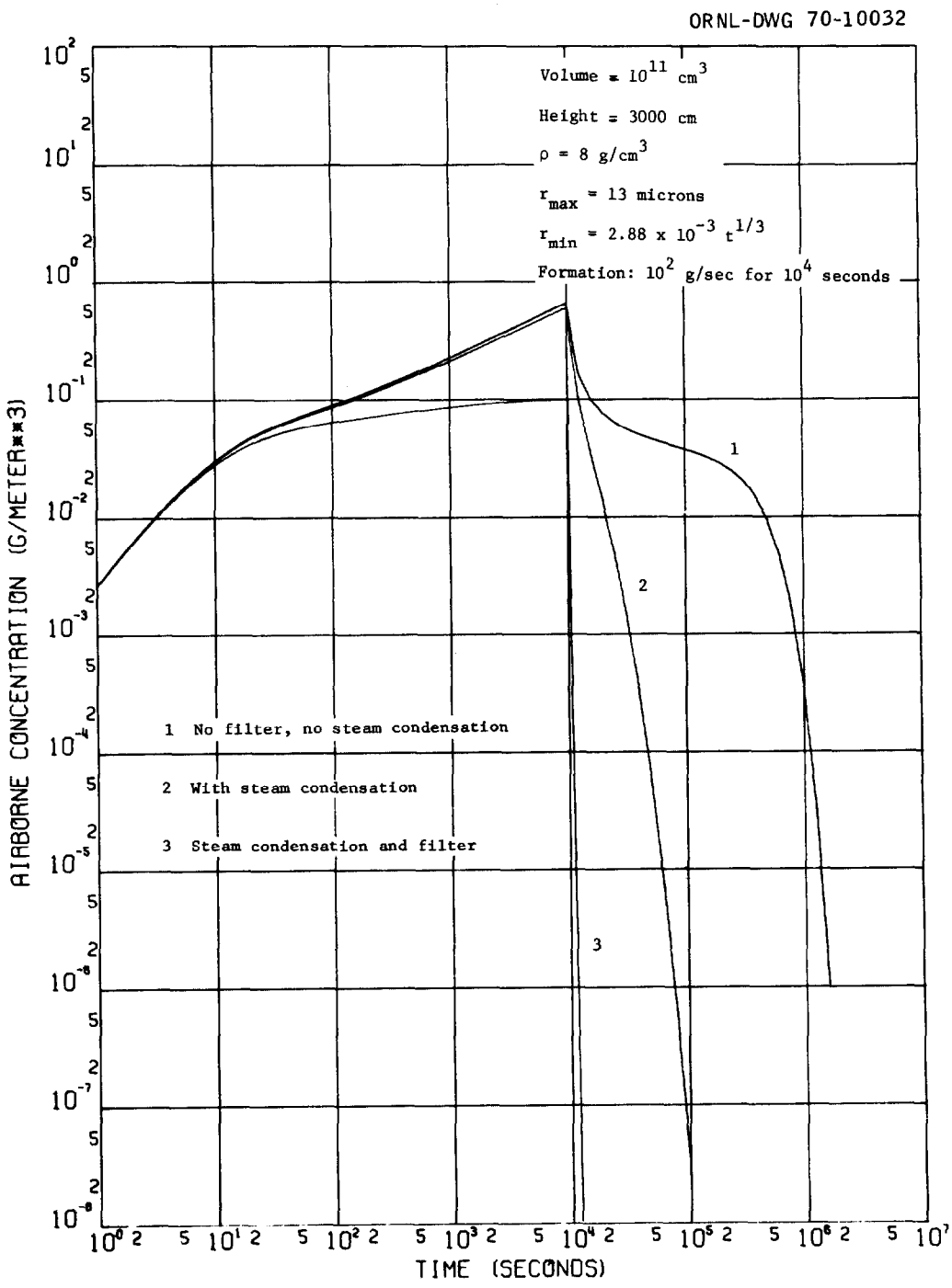
So now we have shown that condensing steam will function to deposit particles; the steam will condense on the particles; the latent heat of condensation will sweep particles toward cold surfaces. The growth of particles due to steam condensation will enhance settling. We have estimated the effect of deposition by thermophoresis on the concentration-time behavior. We cannot now estimate the effect of enhanced settling due to the growth. So we have partially accomplished the third task.

By way of summary, we now present estimates of concentration vs time under conditions expected in a severe water-cooled reactor accident. These are shown on Fig. 18. It was presumed, here, that the maximum particle volume was 104 μm^3, that the minimum particle volume began at 2×10^{-8} μm^3 and grew according to Eq. (7).



EFFECT OF STEAM CONDENSATION

Figure 17



ESTIMATION OF SMOKE CONCENTRATION IN A CONTAINMENT

Figure 18

The particle density was taken to be 8 g/cm^3 (an approximate average between the densities of UO_2 and ZrO_2 and stainless steel oxide). The containment height was taken as 30 meters, the volume as 10^5 cubic meters. 10^6g of aerosol particle substance was presumed to form at a constant rate during 10^4 seconds.

In one estimate (i.e., one curve on Fig. 18) no removal by steam condensation or recirculating filters was assumed. In the second case steam condensation was included; it is suggested that one should be able to depend on steam condensation where containment sprays function because these would ensure a good distribution of cool, condensing surfaces, i.e., the spray drops. The third case also included the effect of a recirculating filter of 100% efficiency with a flow of 20,000 CFM (10^{-4} containment volumes per second). It is clear that both the steam and the filter would be helpful.

To summarize further, we have presented results which demonstrate that the size distribution function given in Eqs. (1-4) does describe aerosols of interest in nuclear safety. We have presented some information regarding the dependence of V_{min} on environment; aerosol age seems to be the most important variable. The effect of the increase in V_{min} with time on the concentration-time behavior was estimated. The steam condensation experiments demonstrated that particle deposition occurs by thermophoresis in condensing steam. Particle growth and enhanced settling also must occur in condensing steam. The effect of deposition by thermophoresis on the concentration-time behavior was estimated. Finally, some estimates of aerosol particle concentration vs time in water-cooled reactor containments were presented.

It is important finally to point out some areas of understanding that are lacking yet may be important. Very little has been done[13] with regard to estimating the amount of smoke possible; further consideration of the vaporization, nucleation and transfer out of the primary vessel could well indicate ways to prove that the greatest possible amount of smoke in water-cooled reactor accidents is quite small. The fraction of the iodine adsorbed on particles, the fraction of that which is reversibly adsorbed and the conditions at which desorption will occur cannot now[14] be estimated with confidence. We mentioned above that particle growth in condensing steam will enhance settling; we cannot now estimate how much. Also the change in size distribution (i.e., increase in V_{min}) with time needs to be considered further. In a system where fresh smoke is being continuously generated (for a period of time) into a large containment volume the fresh smoke will presumably have a V_{min} similar to that of the fresh smoke described above but it will be mixing with older smoke. We cannot now say with any certainty what the resulting size distribution will be.

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DISCUSSION

BAURMASH: The question I had was that you said that the maximum concentration you used on Smokelear was about 5×10^{-3} gr/cm^3. Is this correct?

DAVIS: That's the highest concentration that we employed in our experiments.

BAURMASH: The reason I asked the question is that it sounds like the experiment is in the Browning agglomeration regime of particle concentration. I was wondering how good Smokelear is on high concentration experiments.

DAVIS: Actually the data I used in the demonstration of agreement between Smokelear calculated values and data, was your data.

BAURMASH: That was a low concentration, and that's why I'm asking the question. Have you gotten any data from higher concentration runs?

DAVIS: The highest concentration (in your data which we employed) was 10 gr/m^3. These are data that you reported two years ago, it's for UO_2 aerosol. I think the difficulty here is a mixup between our experimental and our calculated efforts. We dealt experimentally with fairly low concentrations. We have considered in our calculations higher concentrations; indeed any data we could get a hold of.

LARGE SCALE AIR CLEANING TESTS IN THE CONTAINMENT SYSTEMS EXPERIMENT[a]

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ABSTRACT

Results of five CSE experiments in which an internal recirculating air-cleaning loop was run in steam-air atmospheres are presented. The loop was composed of a commercial sized heat exchanger, demister, filters and charcoal beds. Variables investigated were loop flow, atmospheric temperature, component arrangement and aerosol source term. The observed removal of iodine, cesium, uranium, and methyl iodide from the typical water cooled reactor post-accident atmospheres is compared to a model which assumes a well mixed gas space, for which:

$$C/C_o = e^{-\left(\frac{EF}{V} + \frac{kA}{V}\right) t}$$

where,

- C = gas space concentration at time t
- C_o = gas space concentration at time loop starts
- E = loop filter efficiency
- F = loop flow rate
- V = vessel volume

(a) This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1)-1830 with the Battelle Memorial Institute.

(b) Currently with WADCO Corporation, a subsidiary of Westinghouse Electric Corporation, Hanford Engineering Development Laboratory, Richland, Washington, 99352, and under AEC Contract AT(45-1)-2170.

k = natural processes removal coefficient

t = time after loop starts

A = surface area for natural processes

This model applied with E = 0.8 for methyl iodide and E = 1 for other airborne species for about two hours, at which time the removal rates decreased.

Introduction

Current philosophies in the nuclear power reactor safety field are to protect the environs from the consequences of a "loss of coolant" accident by use of engineered safety features. The increasing reliance on the use of these engineered safety systems such as filters, absorbers and sprays requires the performance of such systems to be of demonstrated reliability and effectiveness.

Many reactor plants use air filtering systems to reduce the possible fission product leakage from the reactor containment vessel. This may be accomplished in two ways. The system can reduce the airborne concentration of vaporized fuel and fission products by filters and absorbers. The same system can cool the containment vessel atmosphere, condensing the steam and thereby reducing the pressure within the containment vessel. As a result the leakage rate will be reduced.

A series of tests have recently been completed where the behavior of air cleaning systems was measured in the Containment Systems Experiment (CSE). The CSE tests evaluated the removal of simulated fission products from the containment atmosphere over a likely range of post-accident conditions. Commercial-sized air cleaning components were tested in the internal recirculating loop.

The main objective of these tests was to observe the effect of loop operation on the containment atmosphere. Individual loop components and some combinations have been thoroughly studied and their expected performance is known (1,2,3,4). The tests described here were done with a complete loop of full sized components operating in a range of accident atmosphere conditions and iodine concentrations.

Facilities

Containment Vessel

The CSE containment vessel, a 1/5 linear scale model of a typical power reactor containment arrangement, is 67 feet high and 25 feet in diameter. The vessel is arranged so that either PWR or BWR containment schemes can be tested by proper use of the dry well or wet well

interior structures, which can be seen in Figure 1. For the tests being described, the dry well lid was left in the open position, and the wet wells were closed off. The vessel is equipped with air and liquid sampling systems and is instrumented for temperature and pressure measurements. The main vessel parameters are listed in Table I. Additional details can be found in References 5,and 6.

Air Cleaning Loop

The air cleaning loop is an internal recirculating type, assembled of modular stainless steel units, 3 feet in diameter, and is located across the open dry well. Each unit of the loop contains a gasketed bulkhead to mount a component, such as a filter or charcoal bed. Each module also provides connections for samplers and instrumentation. These sections can be disassembled for component changes. The loop flow was measured with a calibrated orifice plate and manually controlled with an adjustable damper at the blower outlet. The air exhausted at the 30 foot level through a stack. This is typical of many reactor buildings in that cross flow is produced to aid mixing within the vessel. A 10 HP motor and belt driven blower provide air movement. Figure 2 shows the loop in place through the open containment vessel door.

The loop components were selected to be typical of reactor clean up systems. Although many different arrangements are in use or are planned, most use some or all of the components listed: (7)

- Heat exchanger
- Demister® type of moisture separator
- Prefilter
- High efficiency particulate air filter (HEPA)
- Activated charcoal beds

The CSE loop uses nominal 2 ft x 2 ft commercial-sized components operating at 1000 cfm (measured at containment conditions of temperature and pressure). This results in about 2.8 air exchanges per hour in the main room, which is typical of many containment air cleaning systems, although some operate at higher turnover rates.

The heat exchanger is a finned copper tube unit manufactured by Aerofin. The fins are 3/4 in. high, 4 per in. crimped on 1 in. O.D. tubes. The face area is 4.1 sq ft with an estimated total outside heat transfer surface of 163 ft^2. Water flow through the unit was adjusted to provide a steam-air atmosphere ΔT of about 2°F or 3°F as it passed

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AIR CLEANING LOOP AND SAMPLING SYSTEM IN THE CSE VESSEL

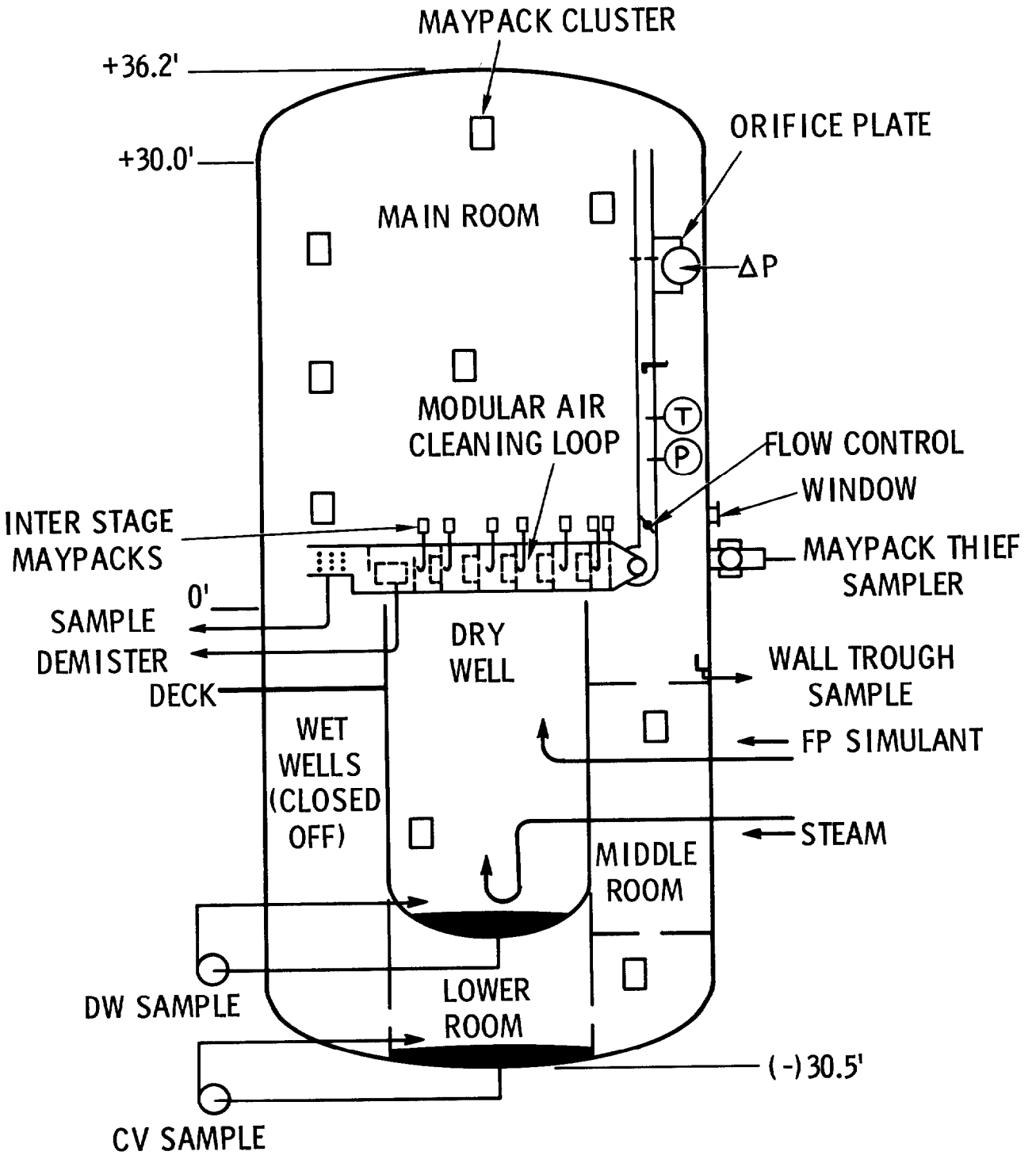


FIGURE 1

TABLE I

Physical Conditions Common to All Filter Loop Experiments

Volume above deck including Drywell	21,005 ft^3	595 m^3
Surface area above deck including Drywell	6,140 ft^2	569 m^2
Surface area/volume	0.293 ft^{-1}	0.958 m^{-1}
Cross section area, main vessel	490 ft^2	45.5 m^2
Cross section area, Drywell	95 ft^2	8.8 m^2
Volume, middle room	2,089 ft^3	59.0 m^3
Surface area, middle room	1,363 ft^2	127 m^2
Volume, lower room	3,384 ft^3	96 m^3
Surface area, lower room	2,057 ft^2	191 m^2
Total volume of all rooms	26,477 ft^3	751 m^3
Total surface area, all rooms	9,560 ft^2	888 m^2
Loop Intake ℄ Elevation	+1.3 ft	0.4 m
Stack Discharge Elevation	+31 ft	9.4 m
Surface coating	All interior surfaces coated with phenolic paint[(a)]	
Thermal Insulation	All exterior surfaces covered with 1 in. fiberglass insulation[(b)]	

(a) Two coats Phenoline 302 over one coat Phenoline 300 primer. The Carboline Co., St. Louis, Missouri.

(b) k = 0.027 Btu/(hr) (ft^2) (°F/ft) at 200°F, Type PF-615, Owens-Corning Fiberglas Corp.

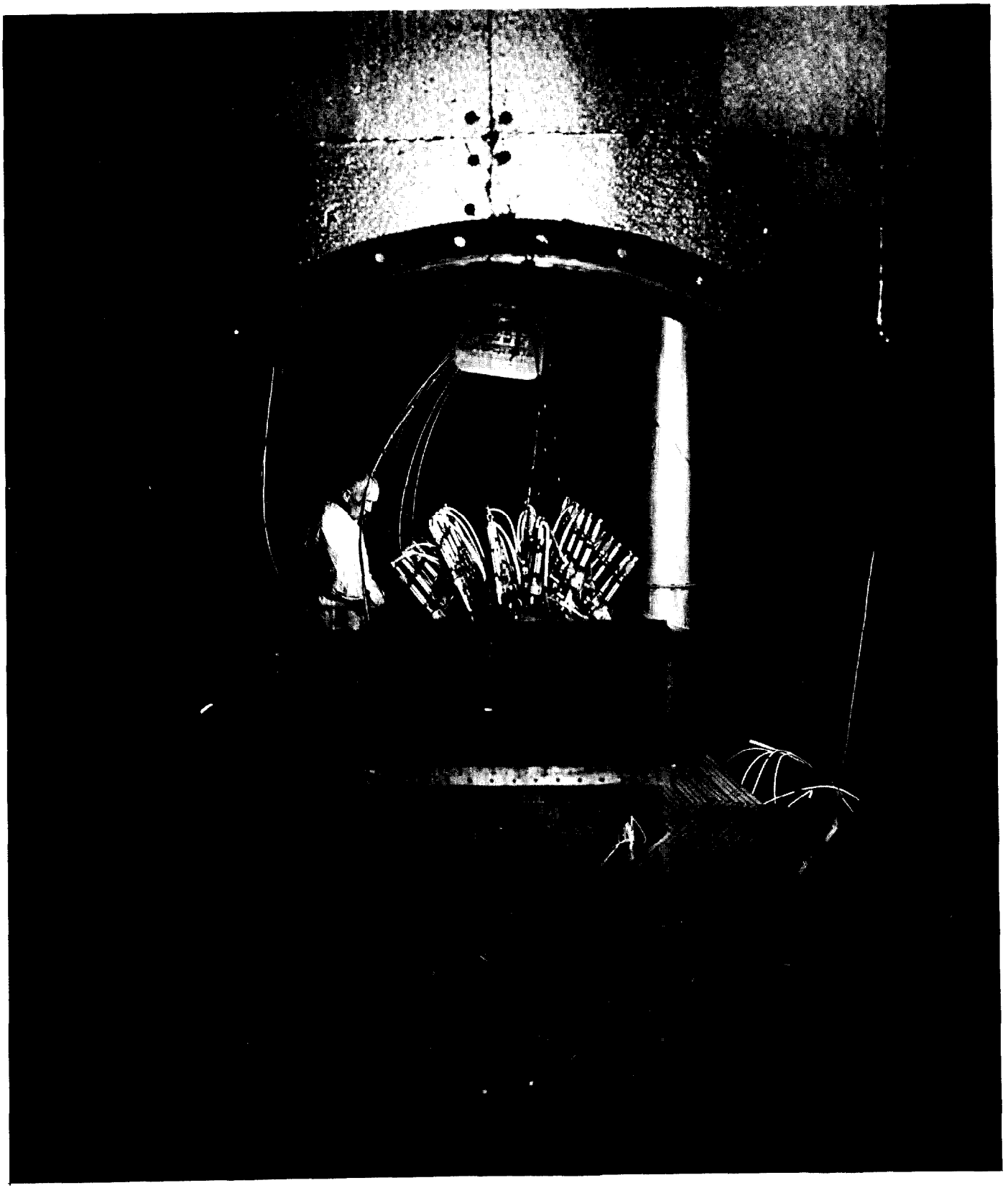


FIGURE 2. VIEW OF AIR CLEANING LOOP SHOWING INLET HEAT EXCHANGER, INTERSTAGE SAMPLERS AND EXHAUST STACK

through the heat exchanger. About 30,000 Btu/min of heat were removed during typical conditions.

The Demister unit was of stainless steel wire-Teflon yarn knitted construction, type 321-SR, 2 in. thick, made by Otto H. York Company. Two arrangements were used. Up flow at 1000 cfm through an 18 in. diameter unit and horizontal flow at 1800 cfm through a 2 ft x 2 ft square unit. Baffles were placed upstream of the 18 in. diameter unit to aid droplet removal by impingement. No such baffles were provided for the horizontal flow arrangement. Any water draining from the units was sampled.

Prefilters are commonly used in filter trains to protect the absolute filters from mechanical damage and from needless loadings of lint and dust. Such a prefilter was provided in the CSE tests just upstream of the HEPA filter, in all except one run. An expanded aluminum mesh filter 2 ft x 2 ft, 2 in. thick, type BAA2-45 supplied by American Air Filter Corporation was used.

The high efficiency particle air filter was of construction suitable for use in the moist high temperature environment. This filter was purchased from specifications given by Burchsted and Fuller (7) in their Appendix A. The filter had a waterproof glass medium, cadmium plated steel frame, aluminum separators and neoprene bond and gaskets. The filters were tested by the USAEC Western Quality Assurance Station after arrival on the site.

The pleated type charcoal beds were type FE manufactured by the Barnebey Cheney Company. The stainless steel frames were loaded with about 50 pounds of 8-16 Tyler mesh (2.36 to 0.99 mm) iodine impregnated charcoal, BC type 727. Each bed is one inch thick.

The three charcoal beds are arranged in series with the full flow passing through each. The face velocity is higher than normal, but use of 3 in. of total bed depth rather than 2 in. compensates for this, and the residence time is 0.2 seconds which is the time recommended for methyl iodide removal.

Sampling Systems

Both the containment vessel and the loop are equipped with Maypacks for sampling the airborne fission product simulant concentration during the tests. The CSE Maypacks (8) are arranged in clusters of 12 and are suspended throughout the vessel so that the average concentration as well as any concentration differences in the vessel can be measured. Ten clusters were used in these tests.

The CSE Maypack differentiates between particles and various forms of iodine by use of:

- Particulate filter
- Silver plated copper screens
- Charcoal loaded filter
- Charcoal bed (unimpregnated)

Corrections have been developed to adjust for the elemental iodine which may be retained on the filter and for methyl iodide on the charcoal paper. A charcoal bed in dry ice serves as a backup to trap any penetration of iodine forms through the Maypack.

After each section of the loop, similar Maypacks are used to sample the atmosphere at up to five times during a run. A sixth loop Maypack at each section is used as a blank.

Samples are taken of all liquid streams and also through special sample ports for particle sizing. Deposition coupons are used to measure the amount on surfaces and to aid in constructing material balances after each run. The amount of material in each loop section is determined after each run by sampling or by decontamination of the sections.

Simulant Generation

The CSE does not use actual fission products in the representation of the loss of coolant accident but uses carefully selected simulants. The validity of simulant use in containment studies has been documented. (9,10)

The general method has been the high temperature vaporization of the iodine, cesium and uranium oxides in individual furnaces. These are blended together and injected into the containment vessel. Coleman (11) treats the generation and analysis in detail. The material is radioactively traced before release, and sample analysis was by gamma couting (I^{131} Cs^{137}) or alpha counting (U^{238}).

Design base accident concentrations of iodine were obtained for all runs (about 100 mg/m^3). Methyl iodide was released in concentrations of 5% of the iodine content.

Test Conduct and Conditions

The five tests made covered a range of conditions and loop arrangements of interest to reactor containment systems designers. Table II lists the main features of each run.

TABLE II

Summary of Air Cleaning Run Conditions

	CSE Run Number				
	A-13	A-14	A-15	A-16	A-17
Date	8/19/69	10/14/69	11/11/69	1/20/70	3/10/70
Temp.°F	96	250	250	245	246
Press.,psia	14.5	48	48	48	48
Components in order of flow	Prefilter HEPA (3) Char Bed	Heat Exchange Demister Prefilter HEPA (3) Char Bed	Prefilter HEPA (3) Char Bed	Heat Exchange Demister Prefilter HEPA (3) Char Bed	Heat Exchange Demister HEPA (3) Char Bed
Flow Rate, cfm	1,000	1,000	1,000	1,000	1,850
Turnover/hr (main room)	2.8	2.9	2.8	2.8	5.2
Heat Exchange ΔT, °F	None	2	None	2	1
Aerosol Release, min	10	10	10	120	10
Initial Main Room Conc., mg/m^3					
I	105	100	100	11[a]	105
CH_3I	5	5	5,5[b]	2.5[a]	6
Cs	7	3	2.6	9[a]	2.5
U-Zr	4	1.6	01.6	0.1[a]	1.5

(a) Max. Conc. reached. Loop operated during release.

(b) Two methyl iodide releases were made.

The general sequence of the test was very similar for all runs and proceeded as follows:

- Loop was loaded with new components (Demister was decontaminated and reused)
- Loop tested for leaks and CH_3I and DOP penetration
- Maypack samples installed in vessel
- Vessel leak tested
- Vessel brought to test conditions with power house steam
- Simulants released
- Sampling started
- Loop flow started, and test conditions maintained as required by run plans
- Vessel cooled and loop flow stopped
- Loop DOP tested; samples recovered and analyzed
- Loop disassembled; components removed for sampling
- Vessel decontaminated by additional steaming
- Final material balance made

With the exception of Run A-16, the loop was started 30 or 40 minutes after the aerosol release. This allowed samples of the atmosphere to be taken so that the initial concentration could be determined, so that the extent of main room mixing could be determined, and lastly, so that the natural process removed rates could be determined. Only by allowance for the natural removal can the net effect of the loop be properly assayed.

The runs typically lasted two days, one day at steady temperature and pressure and one day of decreasing temperature and pressure. Loop flow and cooling water flow to the loop were maintained constant during the cooldown portion of the run.

Results

Main Room

The primary purpose of the air cleaning loop is to remove the vaporized fuel and fission products from the containment atmosphere. Because of this, special emphasis was placed on the sampling of the atmosphere within the CSE vessel.

The removal of material by a loop operating in a well mixed gas space can be expressed as

$$C/C_o = e^{-\left(\frac{EF}{V} + \frac{kA}{V}\right)t} \qquad (1)$$

where the terms are as previously defined. EF/V is the removal constant due to the loop operation and kA/V is the removal constant due to natural process such as deposition and reaction on surfaces. Hence,

$$C/C_o = e^{-(\lambda_L+\lambda_N)t} = e^{-\lambda_T t} \qquad (2)$$

The natural processes can be evaluated before the loop operation starts (F=0), and the net effect due to the loop only can be deduced by correcting the observed removal when the loop is operating:

$$\lambda_L = \lambda_T - \lambda_N \qquad (3)$$

The observed effect of the loop operation is clearly shown in Figures 3 and 4, where the loop startup markedly reduces the concentration half time for both iodine species and the particles. This behavior is typical of that observed in all the runs.

A summary of the observed simulant removal in the CSE tests is given in Table III for times soon after the loop startup. In this table, the observed concentration half times are given for the various airborne forms. The average of the corrected half times can be compared with the expected early half time given by t 1/2 = 0.693 V/F.

The overall agreement is good, A notable feature of this table is that except for Run 16, elemental iodine is removed faster than predicted. This possibly is due to enhancement of the natural removal by the loop flow increasing the convection currents within the vessel. The effect of atmosphere, temperature, and deletion of the moisture eliminator is small considering the value obtained for Runs 13, 14, and 15. The effect of flow was as expected. Run 17 shows the faster removal obtained, although at some decrease in efficiency. The removal obtained during Run A-16 is evidence that the source term need not affect the loop operation.

This rate of removal does not continue indefinitely, however, but decreases after several vessel volumes have passed through the loop. This decrease is due to desorption from paint and liquid surfaces acting as sources. Cesium, however, is not expected to desorb, and the high removal persists for longer times than for iodine.

The longer term measure of the loop effectiveness might be the final concentration in the vessel. Table IV gives the concentration in the gas at 24 hours as a fraction of the concentration present in the main room when the loop started.

IODINE CONCENTRATION IN THE MAIN GAS SPACE - CSE RUN A17

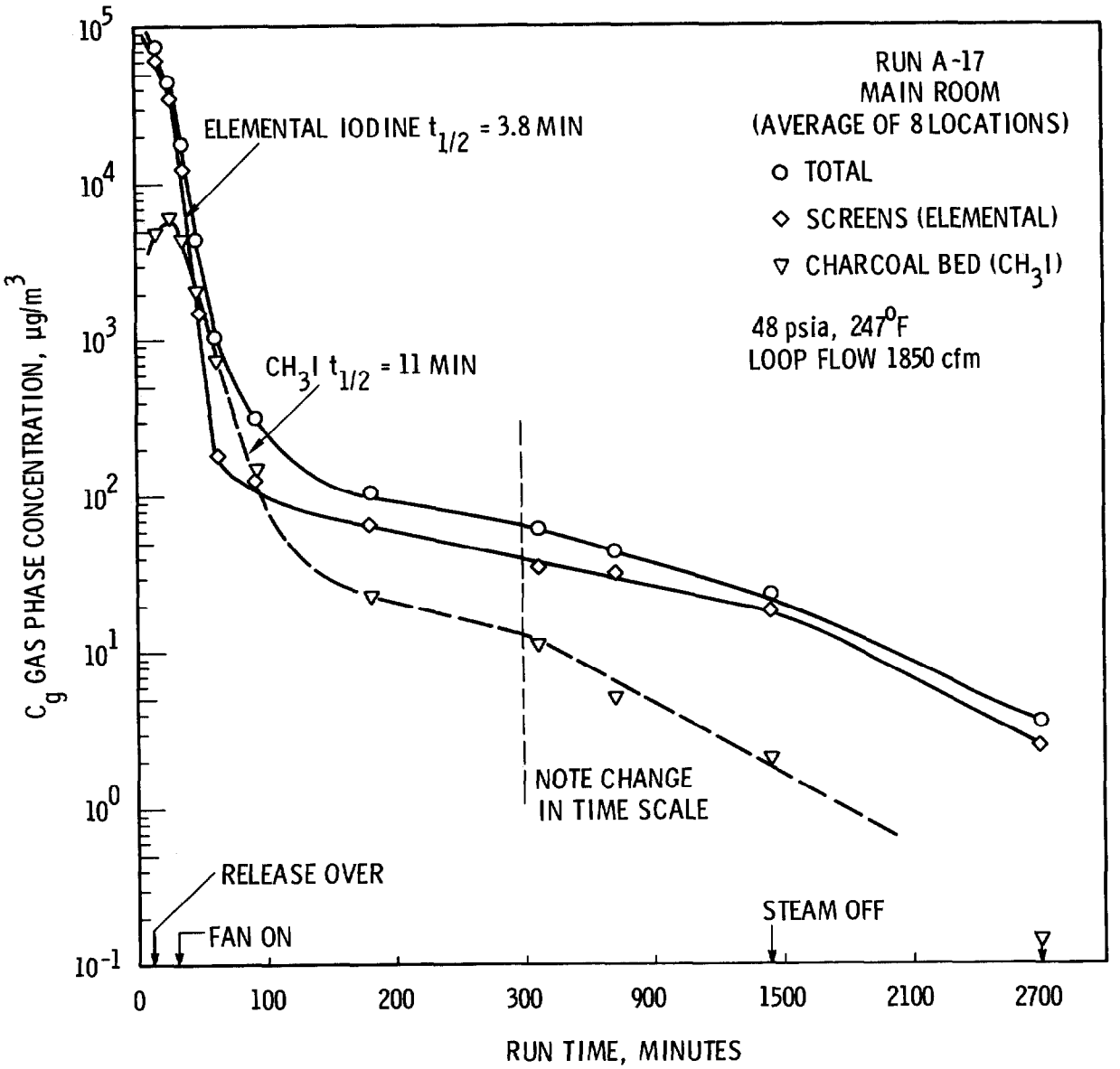


FIGURE 3

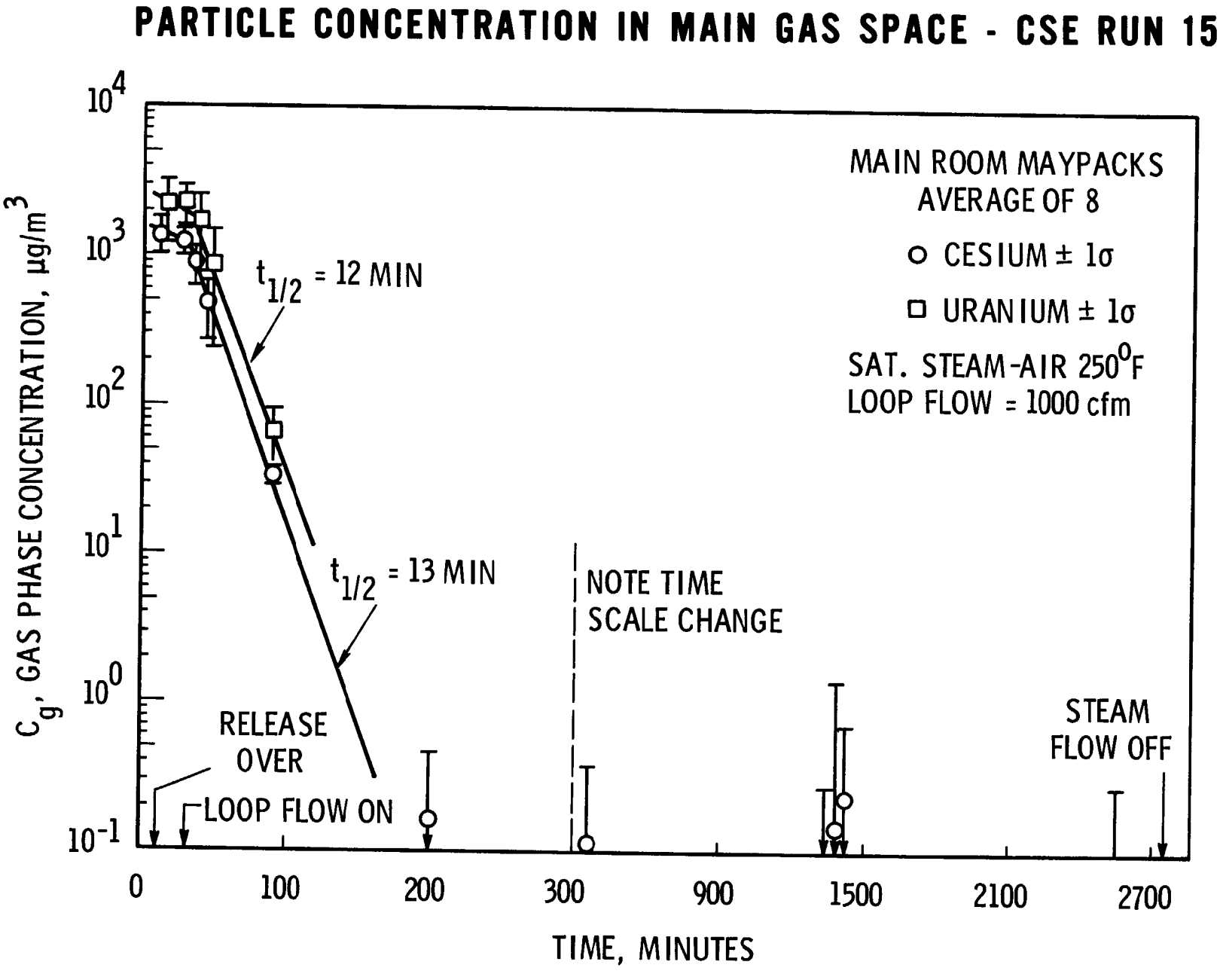

PARTICLE CONCENTRATION IN MAIN GAS SPACE - CSE RUN 15
C_g, GAS PHASE CONCENTRATION, µg/m^3
10^4
10^3
10^2
10^1
10^0
10^{-1}
MAIN ROOM MAYPACKS
AVERAGE OF 8
○ CESIUM ± 1σ
□ URANIUM ± 1σ
SAT. STEAM-AIR 250°F
LOOP FLOW = 1000 cfm
$t_{1/2}$ = 12 MIN
$t_{1/2}$ = 13 MIN
NOTE TIME
SCALE CHANGE
RELEASE
OVER
LOOP FLOW ON
STEAM
FLOW OFF
0
100
200
300
900
1500
2100
2700
TIME, MINUTES

FIGURE 4

TABLE III

Summary of CSE Air Cleaning Results

Early Concentration Half-Times, $t_{1/2}$, minutes

Run	13			14			15			16			17		
	N	N+L	L	N	N+L	L	N	N+L	L	N[a]	N+L	L	N	N+L	L
Elemental I	26	7.3	10.1	16	4.9	7.1	27	6.0	7.7	15	7.3	14.3	13	3.8	5.4
"Particulate" I	50	11.0	14.1	27	9.0	13.5	27	10.3	16.7	36	15.0	25.7	25	6.3	8.7
AC Paper	~50	16.0	21.0	39[a]	9.0	11.8	58	10.0	12.5	39	13.7	14.2	20	7.7	12.5
Char Bed	--	15.0	15[c]	--	14.3	14.3[c]	--	14.7	14.7[c] 19[b]	--	16.0	16.0[b]	--	11.0	11[b]
Cesium	62	11.0	13.7	42	12.3	17.4	70	13.0	16.0	47	9.3	11.5	33	7.7	10.0
Uranium	80	14.0	17.0	85	13.0	15.4	70	12.0	14.5	31	16.0	33.0	41	8.3	10.4
Average			15.15			13.25			14.4			19.1			9.7
Expected[e], min			14.8			13.5			14.6[d]			14.2			7.9
Ratio: Exp./Calc.			0.98			1.02			1.01			0.743			0.81

N = Natural L = Loop Only

(a) Natural $t_{1/2}$, not measured this run. Ave. of 8 CSE runs under similar conditions used.

(b) Bed moisture equilibrated.

(c) "Dry" Bed.

(d) Excludes (c) above.

(e) $t_{1/2} = \frac{0.693\ V}{F}$, F includes steam condensed. Assumed loop efficiency = 100%.

TABLE IV

Concentration Reduction at 24 hours from Loop Startup

Run		Ratio C_{24}/C_L Total Iodine	Cesium
A-13		2.3×10^{-4}	2.4×10^{-5}
A-14		1.4×10^{-3}	5.9×10^{-5}
A-15		2.4×10^{-3}	$\sim 8 \times 10^{-5}$
A-17		4.6×10^{-4}	3.8×10^{-5}
	Average	1.1×10^{-3}	5.1×10^{-5}

The loop was running at 24 hours for all runs except A-14 where the flow stopped at 16 hours. This did not seem to make much change in the final value reached. This faction of iodine remaining would tend to limit the long term DRF to about 1/f or $\sim$ 1000.

Loop Response

Deletion of the heat exchanger and Demister from the loop during Run A-15 did not significantly reduce the loop effectiveness. The HEPA pressure difference did increase more during the run without the Demister, as can be seen in Figure 5. The total loop ΔP was higher in Run A-16 due to Demister insertion, but the change was less. This change was temporary, i.e., the ΔP decreased to nearly normal values during the DOP test after the run. Some moisture may have carried over to the first charcoal bed, as a small increase in ΔP was noted here also.

The efficiency of charcoal for removal of methyl iodide from the humid containment atmosphere is of interest. The CSE tests were all made under conditions of high humidity, and charcoal heating due to moisture adsorption was measured. Two types of charcoal tests were made to explore the effect of the heating and dehumidification. In some tests, the charcoal was exposed to the steam-air atmosphere first, to allow saturation of the bed with water vapor at the test conditions prior to exposure to the methyl iodide. Good removal of methyl iodide by the iodine impregnated charcoal was observed in either case. Figure 6 shows a typical result of bed heating by moisture adsorption. Table V gives the charcoal efficiencies as determined by the observed rates of change in methyl iodide concentration in the main gas space.

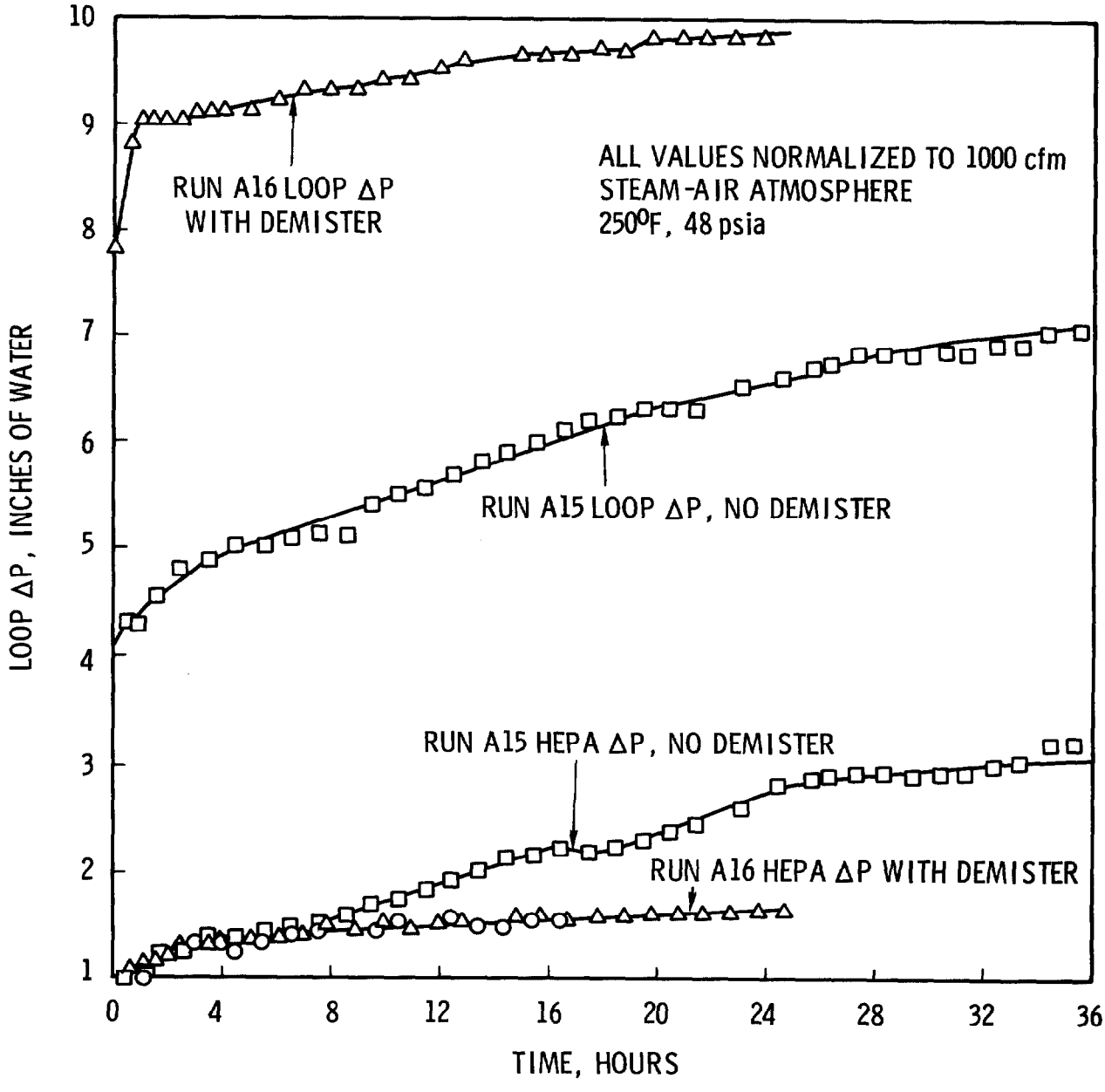
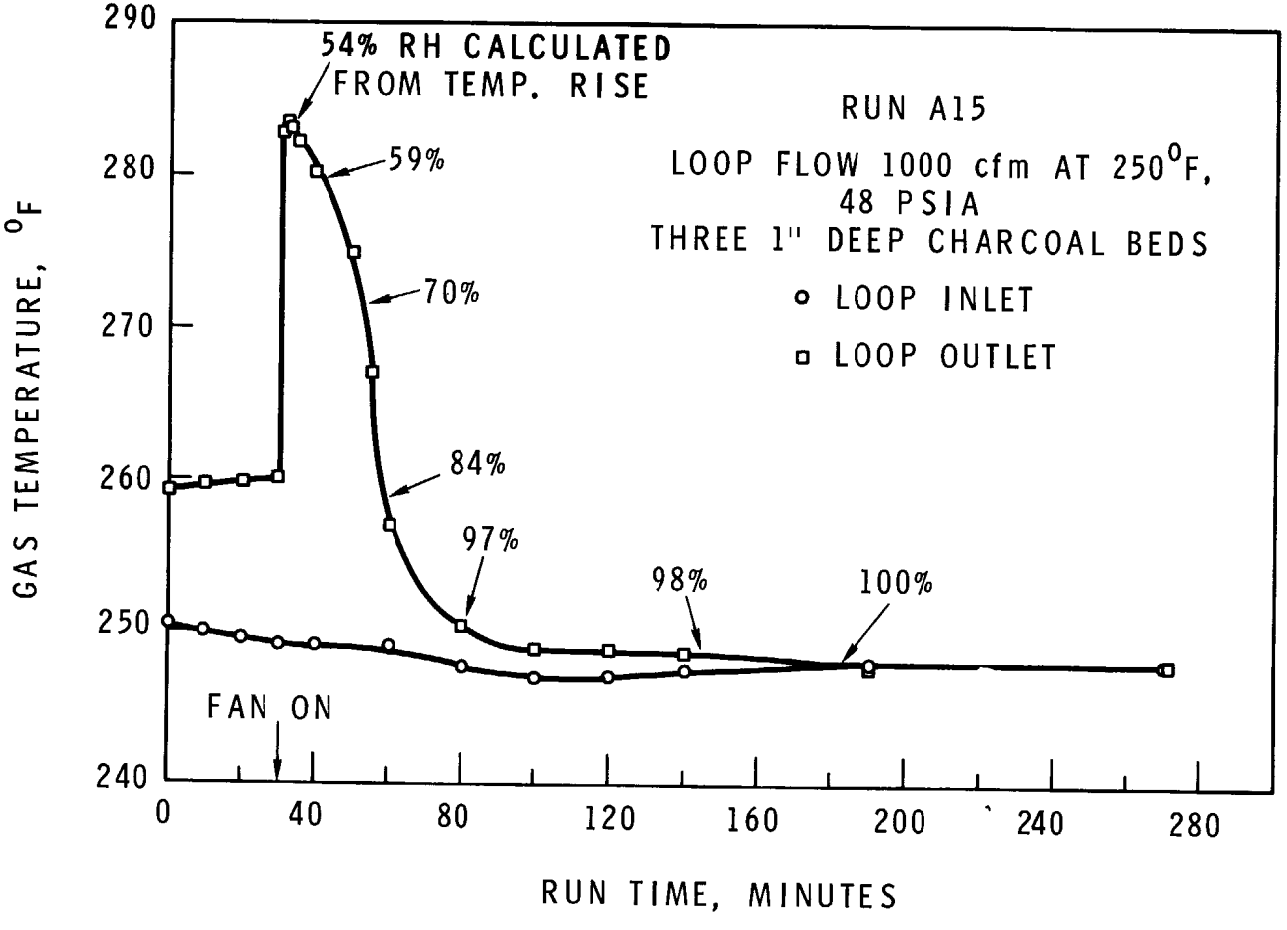


FIGURE 5

CSE FILTER LOOP PRESSURE DROP
SHOWING EFFECT OF DEMISTER



HEATING DUE TO MOISTURE ADSORPTION ON CHARCOAL BEDS, RUN A15

FIGURE 6

TABLE V

Methyl Iodide Removal by Charcoal

Run	Calc[b] $t_{1/2}$ min	Initial Charcoal Moisture Condition: Dry (Self-Heated) $t_{1/2}$, min	Dry (Self-Heated) E[a]	Saturated (No Heating) $t_{1/2}$, min	Saturated (No Heating) E[a]	Integrated Efficiency[c]
A-13	14.8	15	0.99			0.992
A-14	13.5	14.3	0.94			0.9998
A-15	14.6	14.7	0.99	19	0.77	0.9996
A-16	14.2			16	0.89	0.99992
A-17	7.9			11	0.72	0.99993
Average			0.973		0.793	

(a) Efficiency = $t_{1/2}$ observed/$t_{1/2}$ calc

(b) $t_{1/2}$ calculated = 0.693 V/F

(c) Efficiency = Total CH_3I removed/Total CH_3I released.

In the earlier tests, the self-heating of the charcoal prevented bulk condensation in the bed. However, in the later tests using beds which had been previously saturated with moisture, good removal was observed. When the overall removal of methyl iodide during the run is examined, as shown in the last column of Table V, no real difference can be seen. This is of course typical of a recirculating system where good cleanup can be obtained even at lower efficiency per pass.

At the end of each experiment, the loop was again DOP tested for filter leakage and then disassembled, and the components destructively sampled for their iodine and cesium content. Table VI gives the before and after run DOP methyl iodide and duct leakage test results. Operation of the loop in the steam-air atmosphere.did not reduce the filter performance.

Tables VII and VIII give the final distributions of iodine and cesium in the loop. The first one-half of the first charcoal bed retained most of the iodine, as expected, except in Run A-16 where a significant fraction of the iodine was relased as particles. The second and third charcoal beds in Run A-14 seem to have been reversed during sampling.

Significant amounts of cesium were found on the Demister, indicating a filtering efficiency of 50 to 60% at the test conditions.

TABLE VI

Loop Leakage Tests Summary

	Run # A-13	Run # A-14	Run # A-15	Run # A-16	Run # A-17
Pre-Run					
Duct Leakage,%[a]	0.01	0.01	0.02	0.02	0.002
DOP Penetration,%	<0.03	<0.03	<0.03	0.01	<0.03
CH_3I Penetration,%[b]	<0.002	0.02	0.001	0.0003	0.03
					0.04[c]
Post-Run					
DOP Penetration,%	<0.03	<0.03	<0.03	<0.03	<0.03

(a) As % of loop flow at estimated operating ΔP

(b) Penetration in 3 min at ambient temperature and humidity, 1000 cfm

(c) Same as (b) except 1800 cfm

TABLE VII

Iodine Distribution in Loop

	Percent of Total Iodine Found in Loop[a]				
Location	A-13	A-14	A-15	A-16	A-17
HEX	--	0.3	--	9.3	4.0
Demister	--	3.3	--	31.0	0.8
Prefilter	0.2	0.1	0.02	1.3	--
HEPA Filter	8.8	22.0	22.0	28.3	27.0
1st Charcoal Bed 1/2[b]	68.0	42.0	39.0	16.0	41.0
2/2	9.2	19.0	16.0	6.1	13.0
2nd Charcoal Bed 1/2	5.9	0.7	7.9	4.0	5.8
2/2	4.2	0.3	6.2	2.5	4.1
3rd Charcoal Bed 1/2	2.3	8.3	5.4	1.2	2.7
2/2	1.3	4.2	3.9	0.8	2.1

(a) At end of experiment

(b) 1/2 refers to first half of bed thickness; 2/2 refers to second half

TABLE VIII

Cesium Distribution in Loop

Location	Percent of Total Cesium in Loop[a]				
	A-13	A-14	A-15	A-16	A-17
HEX	---	2.5	---	11.0	5.2
Demister	---	62.5	---	51.7	52.0
Prefilter	0.7	0.02	1.5	2.9	---
HEPA Filter	99.3	35.0	98.5	34.4	42.8

(a) At end of experiment

Comparison with Sprays and Natural Processes

A total of 19 large-scale experiments were performed during the CSE fission product transport study, of which only five air cleaning tests are discussed here. Eight spray experiments are reported in References (12,13), and six natural process tests in References(14). All tests were made in the same vessel using the same sampling techniques and under similar conditions. A comparison of the relative effectiveness of the three system types can be made. In Figure 7, the concentration of the total iodine in the main room gas space is plotted for three experiments. In one experiment (A-11), only natural removal effects occurred; in one (A-10) a caustic-borate spray was operated for 24 hours; and in the third (A-14), the air cleaning loop operated continuously for 16 hours, starting at 30 minutes. The time scales have been adjusted to allow for this 30 minute time offset. As expected, the sprays were the most effective at long times. However, after the first half hour, the filter loop was the most effective due to its ability to remove methyl iodide more rapidly than the other two systems.

The real test for effectiveness of removal systems is the reduction in the fraction of fission products escaping the vessel. Assuming that the gas leakage rates were identical in the three tests, the relative amount of iodine which leaked can be calculated by numerically integrating the concentration over the time period of interest. This can be expressed as a "dose reduction factor" (DRF). It is the ratio of the concentration if no reduction occurred to the average concentration during the specified period.

$$DRF_t = \frac{t\ C_{go}}{\int_o^t C_g\ dt} \qquad (4)$$

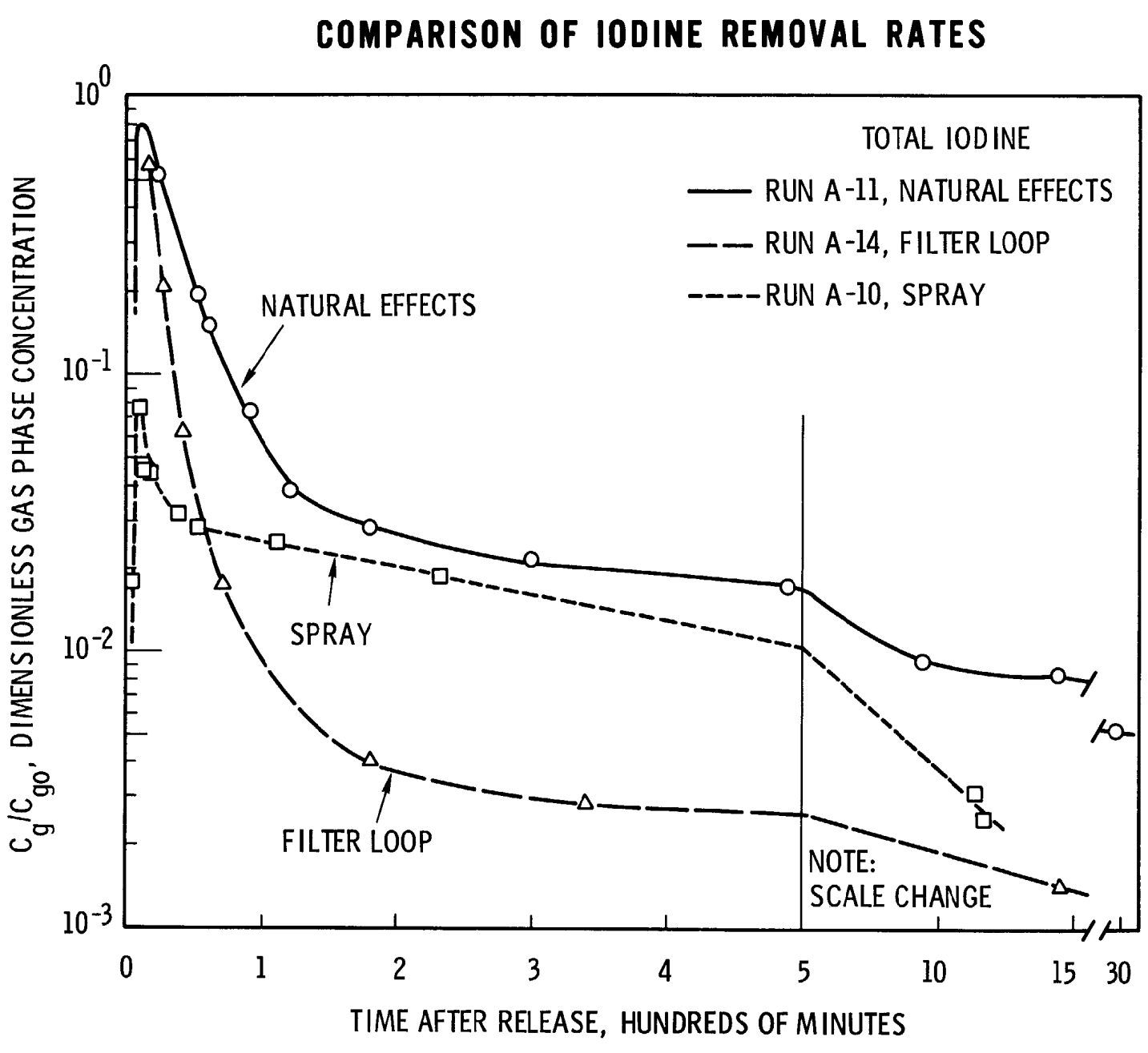

COMPARISON OF IODINE REMOVAL RATES
TOTAL IODINE
RUN A-11, NATURAL EFFECTS
RUN A-14, FILTER LOOP
RUN A-10, SPRAY
NATURAL EFFECTS
SPRAY
FILTER LOOP
NOTE:
SCALE CHANGE
10^0
10^{-1}
10^{-2}
10^{-3}
C_g/C_{go}, DIMENSIONLESS GAS PHASE CONCENTRATION
0
1
2
3
4
5
10
15
30
TIME AFTER RELEASE, HUNDREDS OF MINUTES

FIGURE 7

The 2-hour DRF was 4.2 for natural processes, 8.0 for the filter loop and 30 for sprays. It should be pointed out that natural effects undoubtedly contributed to removal during the spray and filter loop tests. The superiority of sprays diminished with time, and at 24 hours, the $DRF_{24\ hr}$ was 30 for natural, 80 for filters and 100 for sprays.

Conclusions

The main conclusion resulting from this series of tests of air cleaning under accident conditions is that the air cleaning loop performed as designed and was effective in reducing the gas space concentration. Iodine and particles were removed by the loop with an efficiency of 1.0 for two or three hours, after which time the removal efficiency decreased. Methyl iodide removal efficiency by the loopscontaining moisture-saturated charcoal was about 80%. The operation of the loop in the containment vessel appears to have enhanced the natural removal processes for elemental iodine, resulting in faster removal than expected in some cases. The main room gas space remained well mixed in all the tests. Extended duration releases of fission product simulants did not cause any significant changes in loop performance.

The loop components performed well in all the atmospheric conditions tested. Deletion of the heat exchanger and moisture separator caused only minor effects on the filter pressure drop.

The reduction of the gas space concentration for all simulant forms effected by the loop was compared with the performance of caustic sprays. The typical caustic spray system will rapidly remove elemental iodine but will remove methyl iodide only very slowly. A combination system using both filter-absorbers and sprays should be very effective in obtaining large dose reduction factors at short times. Such a combination system, however, was not tested during the CSE program.

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DISCUSSION

WITT: You had one slide showing differential efficiencies of your carbon beds starting in a wet condition and starting in a dry condition that suffered self heating and you had efficiency numbers something like 96%, 73%, and then you had a summation column and you said if you look at the integrated efficiencies at the 99%+ you have a hard time finding a difference anywhere. It wasn't clear to me how you went from the individual bed efficiencies to integrated number like 99. Could you cover that again?

McCORMACK: Yes, the individual bed efficiencies were based on the inlet and outlet concentrations of the beds. Total loop efficiency was based on the removal half time in the main room versus the expected half-time based only on the flow. This gives a per path efficiency. The integrated efficiency numbers that I gave were based on the atmospheric concentration at the end of a run compared to the amount that we had put in the vessel at the start of the run. This is the percent remaining airborne or a fraction remaining airborne, at the end of say a fifty hour run.

KOVACH: Have you found either by direct measurement or by difference anything resembling the hypoiodous acid of Idaho Falls?

McCORMACK: Our may pack doesn't identify HOI as a separate item; we think that it's trapped in our may pack. I didn't fill you in, but one of the may pack components we have is charcoal paper and we think that this may be rich in HOI. So I don't think I have answered your question, but I haven't completely avoided it either.

FICKS: On your methyl iodine runs, what were the approach velocities through the charcoal filters?

McCORMACK: We had standard two by two units and we ran them at 1000 cfm. The approach velocity is about 75 feet per minute under those conditions. This is higher than you would normally use for methyliodide, but we did it because of a design problem that we used three inches of charcoal instead of two to give an equivalent stay time of 0.2 seconds which is the normal value recommended.

FICKS: If you had the same conditions and lowered them down to say the 40 feet per minute that is more or less standard, would you expect to get better results with more residence time or were you satisfied that you achieved the highest efficiency that you could get under the conditions.

McCORMACK: If the residence time had been longer, yes I think the efficiency would have been better.

BURCHSTED: Have you done any work at all on rapid pressure transients and their effect on the components of the air cleaning system?

McCORMACK: No, we haven't. Our pressurization of the vessel is a very slow process, and so there are no transients of any magnitude.

BEATTIE: I assume the charcoal beds were impregnated with potassium-iodide. Is that so?

McCORMACK: Yes, they are impregnated.

FIRST: On the experiments that you ran, you got perceptively better results when the air flow rate was increased or, putting it another way, when there were more air changes per hour. You also mentioned that you had good mixing in the vessel. I wondered if you would tell us how you determined this. I also want to make the comment that it's common knowledge among heating and ventilating engineers that you must get a fairly high number of air changes per hour to get effective room ventilation. I would have thought that the higher results attained with the greater number of air changes reflected this fact. Your statement that you had well mixed gas at all times does not seem consistent with my conclusion.

McCORMACK: The statement on a well mixed main room was based on examination on the individual may packs distributed throughout the main room. We had samples in ten places through the rooms and generally those samples agreed, which made us feel that there were no vast differences in concentrations, say within 20% or so of each other. There were three minute duration samples taken eleven times during a run. Now there may have been instantaneous concentrations within a three minute sampling time. We couldn't see that.

PARKER: Did you observe a natural removal rate for methyl iodide which indicates that methyl iodide is being adsorbed in the paints?

McCORMACK: Yes, we do observe some removal of methyl iodide. I can't give you the number, because I don't have one here with me, but Mr. Hilliard is here and he probably could if you would like it.

HILLIARD: We have made 19 experiments in the CSE and the average half life of the methyl iodide in the experiments where the atmosphere was a steam-air mixture at about 250°F was about 30 hours. This was in absence of engineering safeguards, just by natural deposition on the painted surfaces and by hydrolysis in the steam condensate.

PRESENT STATUS OF THE DESIGN AND TESTING OF RADIOACTIVE FILTRATION INSTALLATIONS AND ASSOCIATED AEROSOL RESEARCH

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ABSTRACT

Present AWRE philosophy on safety aspects of the design of filtered ventilation systems for radioactive process buildings evolved several years ago but is now restated.

Development of the sodium flame test as an in place test is discussed and a method of producing suitable quantities of a stable aerosol for this purpose is described.

For reactor applications the effect of high temperatures on the performance of HEPA filters has been examined and the need for in place testing of units subjected to these conditions is indicated.

An associated aerosol research programme in which the filterability of submicron particles by high efficiency media was investigated is reviewed. Papers have already appeared on this topic and the latest and final one, now in preparation, deals with filtration of plutonium bearing aerosols.

Safety Philosophy

The broad principles of AWRE policy on the design of filtered ventilation systems for radioactive and toxic process buildings evolved several years ago (1). The majority of these systems are intended to deal only with particulate hazards and consist basically of HEPA filters, together with primary collectors of lower efficiency where appropriate. These design principles recognise the dual function of the filtration plants, namely:-

(a) To operate continuously under ambient conditions with maximum overall economy and minimum maintenance requirements; under normal conditions of operation in many cases these systems have to deal with comparatively low levels of contamination, since work on highly toxic materials normally takes place in glove boxes which provide a separate containment barrier.

(b) To maintain integrity under acident emergency conditions. In the event of a fire or incident involving breakdown of these inner containment barriers and the release of radioactive material in aerosol forms the filters would be required to retain very much larger quantities of material. It is important therefore that the HEPA filters should retain their efficiency under the conditions that would occur during such

incidents, particularly high temperature. As a corollary to this the plant must be designed to ensure that during the course of any credible accident the air temperature at the filters does not exceed those which it is known they will withstand.

In practice the second requirement (b) influences the system design and the specification of the HEPA filters to a very large extent. The four principles or guidelines to ensure maximum safety and economy are:-

(1) Use of non-combustible HEPA filters

All filters used at AWRE are purchased against a specification which determines such factors as materials of construction, i.e. material of frame construction, which is normally steel, filter media, i.e. glassfibre paper; this must be of a minimum thickness and weight and must meet a minimum filtration efficiency requirement. The filter itself must be of a design which will withstand heating to a temperature not exceeding 500°C, without loss in efficiency; in some cases it may be necessary to place two HEPA filters in series to obtain the necessary efficiency.

(2) Use of prefilters or primary collectors

Primary collectors such as cyclones or wet scrubbers for very dusty foundry type operations are installed where necessary; more usually glass fibre pad or glass fibre paper type prefilters are installed to give the best economy of operation. Glass fibre mat type pre-filters have been used for some considerable time but recently it has been found that an absolute type filter constructed of glass fibre paper with an initial penetration of between 2% and 5% in fact offers greater economy of operation for use as a prefilter. This is because of reductions in the price of the paper type filters and the gradual increase in the price of the glass fibre mat materials.

(3) Use of a debris arrestor

This is installed in the duct work immediately following the roughing filter or, in some cases, the first absolute filter where more than one is installed. Although the roughing filters and absolute filters are non-flammable and non-combustible it is recognised that flammable dusts can accumulate on them which could possibly be ignited. It has been found experimentally that a forty mesh stainless steel gauze debris arrestor will prevent passage of burning material of the type which could arise from an ignited roughing filter from damaging the final absolute filter in the system.

(4) Isolation or separation of final filter

The final filter or filters in a system are separated from the prefilter and debris arrestor by a minimum distance of, in

some cases, 20 ft of duct work and in other cases a longer distance depending on the diameter of the duct and the airflow through it. This ensures that hot combustion gases or small debris which has passed the arrestor will have cooled adequately before they reach the final filter and so prevent it from being breached.

In assessing the overall safety of the plant, note is taken of the quantities of inflammable material which could possibly accumulate in the ducting or on the roughing filter, its temperature of ignition, heat of combustion, the resulting temperature of the exhaust gases and the dilution and cooling of hot gases by other gas streams from confluent ducts. Limits are also placed on the quantities of inflammable solvents which can be used at work stations.

New plants generally have been constructed in this manner, and where necessary older plants have been modified as far as was found necessary to meet the safety requirements.

Figure 1 indicates the features diagrammatically.

In-Place Testing

Because of the design and method of operation of extract filter plants at AWRE in-place testing is not normally considered necessary. At AWRE the HEPA filters are normally installed inside individual steel canisters which can be pretested before installation and it is then only necessary to instal canisters between headers. The efficiency of the plant as a whole does not, therefore, depend upon gasket seals having to be made each time the filter is installed or replaced. Canisters are of two types depending on application, firstly two or three stage canisters with separate compartments into which the inserts are sealed by a wedge or a roller cam action and can be replaced; and secondly the all-welded construction when the complete canister and filter unit has to be replaced. This later type is normally used where infrequent replacement is necessary.

Figure 2 shows an example of construction involving the use of throwaway all welded canisters for the final stage of absolute filtration preceded by a length of ducting, and the two primary stages of filtration where inserts can be replaced in situ.

Some other UKAEA establishments have employed an in-place test using a Pollak condensation nucleus counter (2) but because of lack of knowledge of the condensation nuclei and their behaviour in filters and filter systems there is some doubt whether the test can be considered suitable for a standard method. Additionally the Pollak counter gives only intermittent readings which means that it is difficult to use with a probe technique for locating leaks.

For some time now efforts have been made involving jointly Harwell, Aldermaston and CDEE Porton to utilise the principles of the sodium flame test for high efficiency filters for an in-place test. In general this work has concentrated on producing more simplified and

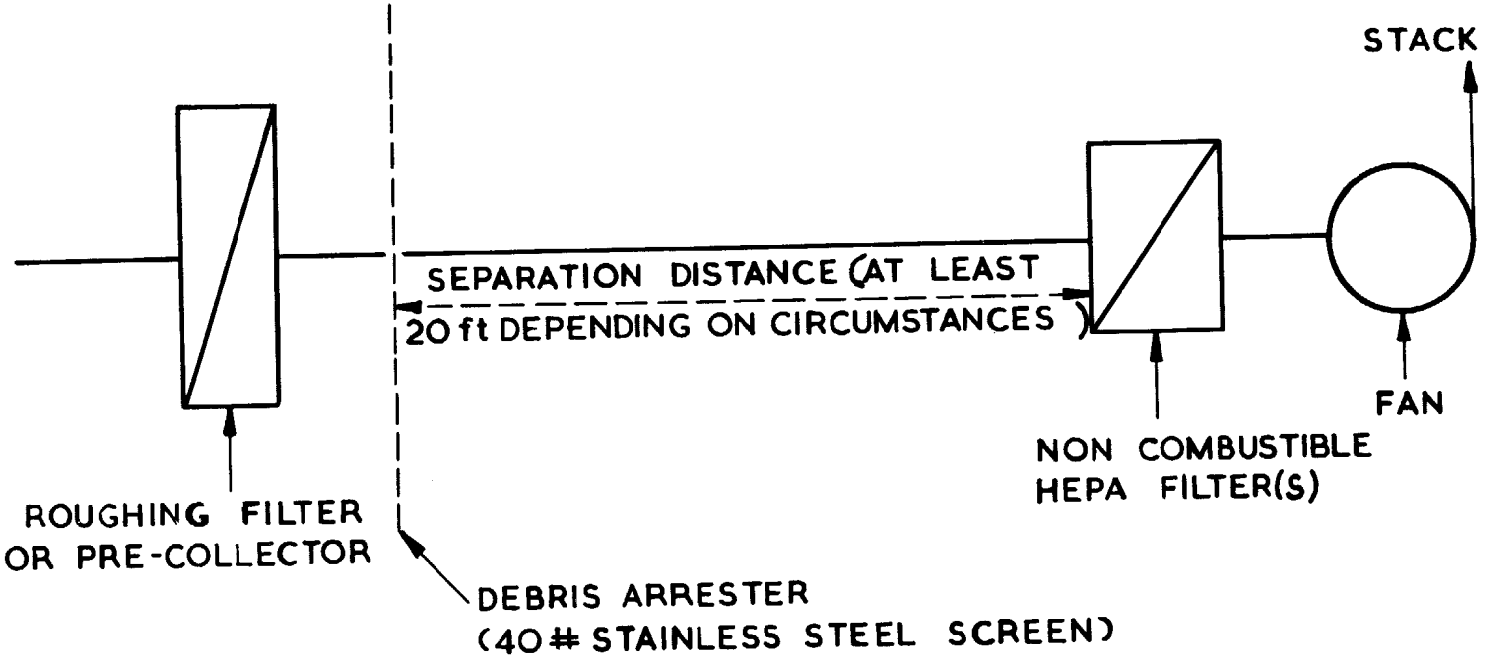

FIGURE I. INSTALLATION OF HEPA FILTERS AT AWRE

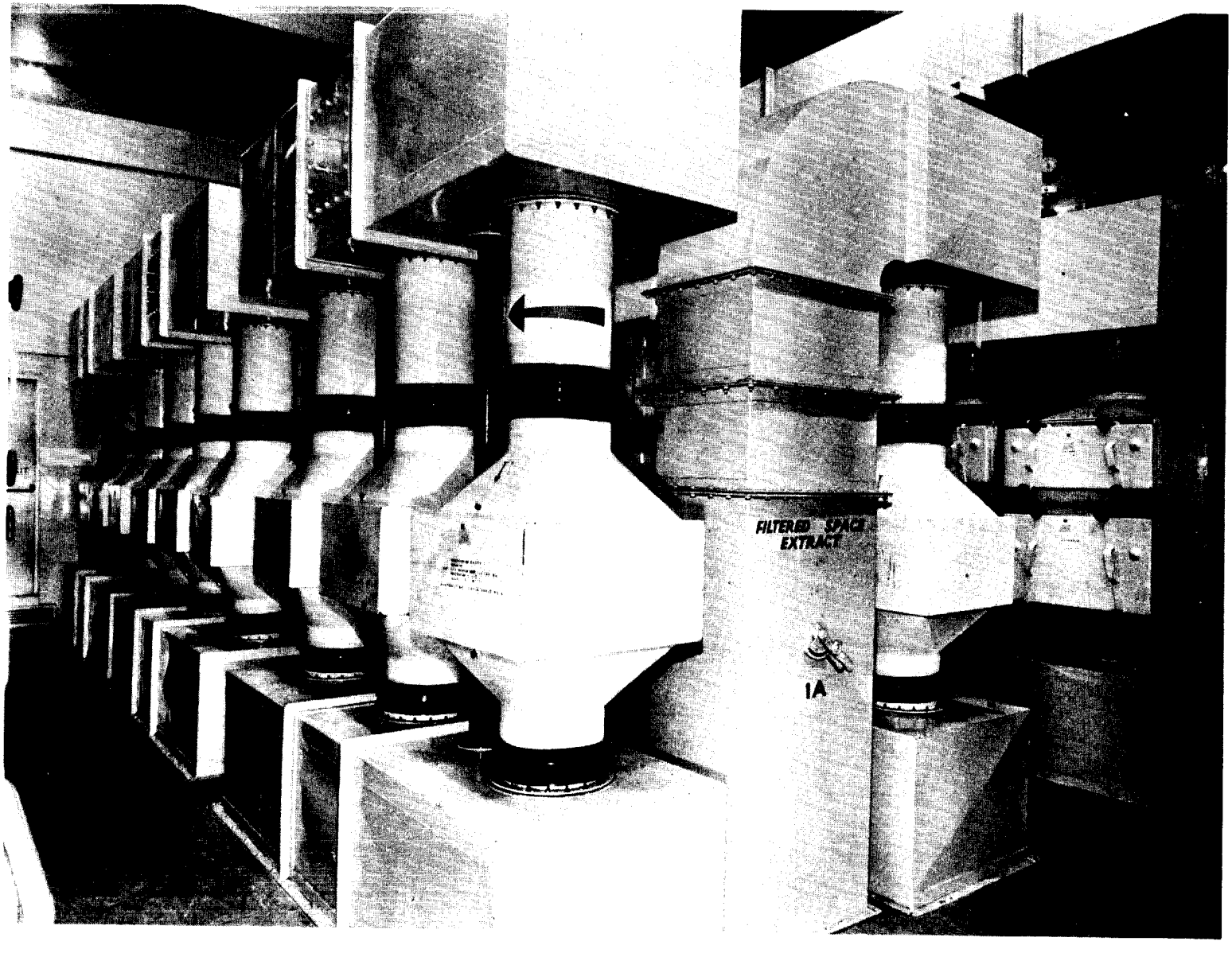

FIGURE 2. INSTALLATION OF HEPA FILTERS IN FUEL ELEMENT MANUFACTURING FACILITY

robust detector systems to replace the present photomultiplier and sensitive galvanometer and the associated high voltage supply. However, none of these developments has offered an alternative method of generating the required quantity of sodium chloride test aerosol using reasonably small and portable equipment and without the need for large volumes of high pressure compressed air.

At AWRE a number of alternative methods of producing a sodium chloride aerosol or sodium containing aerosol which do not necessarily require bulky equipment have been investigated. These include atomisation methods, thermal methods and pyrotechnic methods. The atomisation methods included consideration of possible alternative solvents both organic and inorganic together with the possible use of alternative sodium containing compounds. For example, solutions of sodium nitrite and sodium iodide in anhydrous ammonia were atomised but the dispersion was poor and other severe practical difficulties were also encountered. Thermal methods consisted in principle of heating solid sodium chloride in a gas stream to a temperature such that its vapour pressure was appreciable and then allowing the vapour laden gas stream to cool thus forming an aerosol, but again difficulties were encountered in producing the required output of between 1 and 10 grams per minute. The maximum output which could be obtained by this method was only 50 milligrams per minute. Direct electrical heating of the sodium chloride was also tried but aerosol production rate was extremely erratic and the life of the containment refractory materials was very short because of corrosion and thermal stress. Pyrotechnic methods, i.e. the use of a "smoke candle" giving an output of sodium chloride smoke, were also examined. Generally speaking the requirements of long burning time and large smoke output tended to be conflicting or mutually exclusive since slow burning materials with an adequate burning time burnt with comparactively cool reaction producing little smoke and leaving a large residue of carbon and salt; fast burning combinations giving high temperatures were required to produce more smoke and less carbonaceous residue. The burning times were then too short, however.

Direct Heating of Powdered Sodium Chloride by Burning Gas Mixtures

A steady output of between 1 and 3 grams per minute of a submicron sodium chloride aerosol was obtained by feeding powdered sodium chloride into an oxyacetylene flame. A powder deposition torch was used for this purpose. It was found that, provided the feed rate of salt to the flame was kept at above a certain level of, say, 10 grams per minute, and provided that the gas and oxygen flow-rates were maintained constant, then the output of aerosol remained constant also. Only a proportion of the salt fed was vapourised; the remainder was deposited as a fused cake on the small hearth below the torch. The rate of output could be varied by altering the flame size. The particle size of the cloud obtained was entirely in the range of .01 to 2 microns; the mass median diameter was between .3 and .4 microns.

Used in conjunction with a portable sodium flame detector unit based on the British Standard version, the output from the smoke generating equipment described was used to test a 35000 c.f.m. HEPA filter installation in a process building. The sensitivity of the equipment, as used, enabled penetration values to be determined to the nearest .005%.

The Effect of High Temperature on the Performance of HEPA Filters (3)

Although the filter units used by the UKAEA are required to be of a type that will withstand a single exposure to an environment at 500°C without loss in efficiency, this does not necessarily mean they are suitable for general use in high temperature environments. Tests have been made, therefore, to determine how the filtration efficiencies of two commercially available types of unit were affected by exposure to short term and long term temperature cycling under static and full flow conditions.

Figure 3 shows two HEPA filters being placed in a recirculating hot-air furnace.

The results of the tests indicate a special need for in-place testing of units when they are used at elevated temperatures. For the types of filters examined, which both had constructions avoiding the use of adhesives, it was found that at temperatures up to 200°C there was very little effect on the filter efficiency even after up to 30 heating and cooling cycles and a total exposure time of up to four weeks at the elevated temperature. Up to 300°C, some useful life could be expected providing the number of temperature cycles was not large; an efficiency check after each exposure would be recommended. At 400°C and above, use would not be recommended except for applications involving a single short duration exposure to the high temperature environment, the assumption being that the unit would be replaced after this occurrence.

Aerosol Research Programme

Investigations in which the filterability of sub-micron particles by high efficiency type filter media is examined have been in progress for some time now. The object of the work is to determine filtration efficiency as a function of particle size and gas velocity for the range of particle sizes considered important, i.e. below 1 micron in diameter. Earlier work reported elsewhere (4) with heterogeneous sodium chloride aerosols demonstrated that the size of particle most difficult to remove by filtration was in the range from .1 to .12 microns. The methods used here were to obtain particle size distributions of the aerosol clouds before and after passing through the filter by taking samples with a point to plane electrostatic precipitator and assessing these by sizing and counting from electron micrographs. Subsequent investigations with a different test aerosol material, namely methylene blue with a similar particle size distribution, was carried out using a Goetz aerosol spectrometer to obtain particle size distributions (5). Having established these methods using standard aerosols, i.e. aerosols normally used for filter testing, the technique were extended to measurement of the penetration through filters of radioactive aerosols of high density such as might occur in the extract systems of radioactive processing buildings and facilities. This work is in progress at the moment. For the uranium oxide aerosol work the most suitable method of obtaining particle size distributions was found to be again from the assessment of electron micrographs as in the case of the sodium chloride work. Difficulties were experienced in applying the Goetz spectrometer to this material because of insufficient sensitivity of the radioactive counting methods and the activation analysis methods of detection.

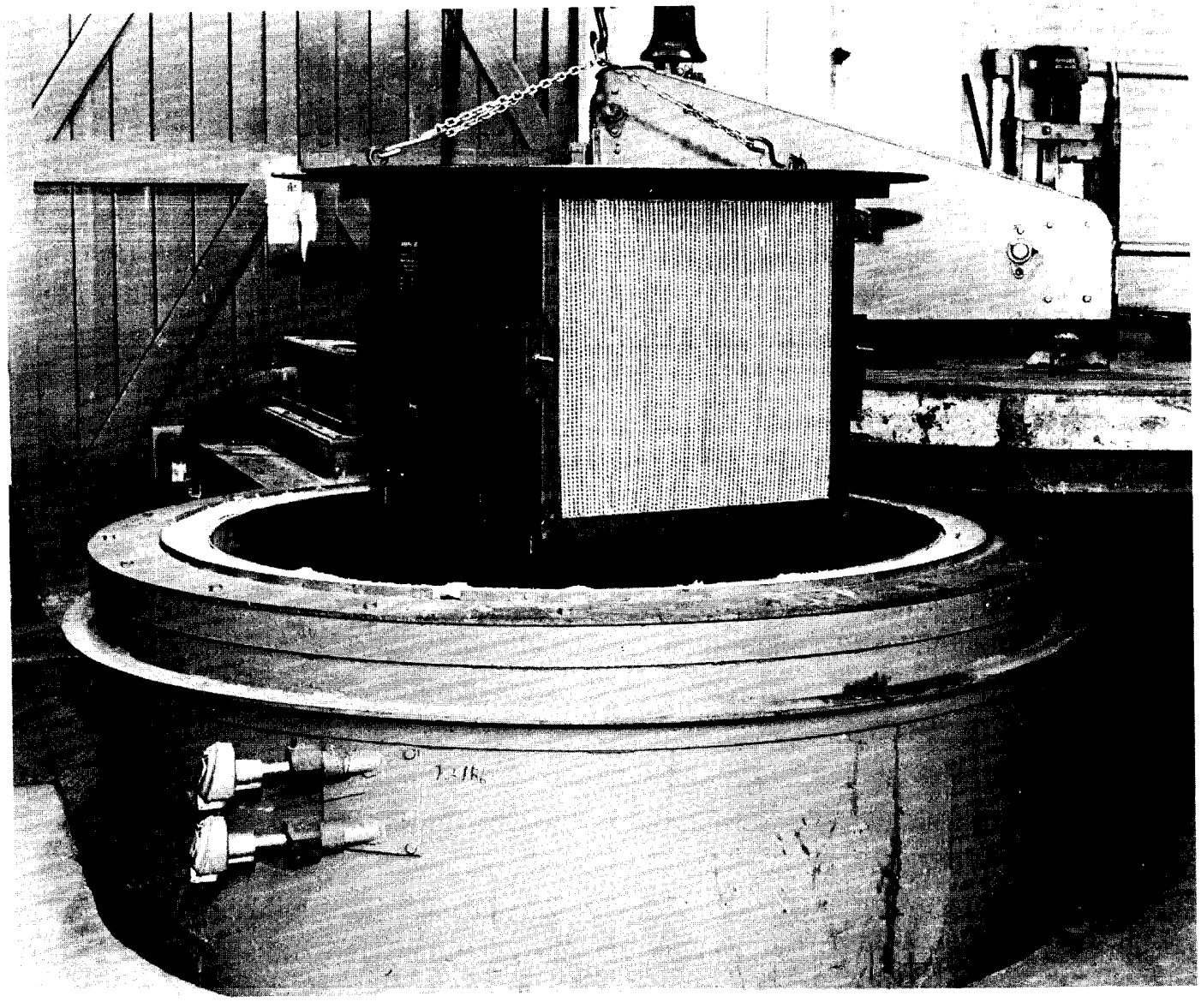

FIGURE 3. ABSOLUTE FILTERS INSTALLED IN RECIRCULATING OVEN

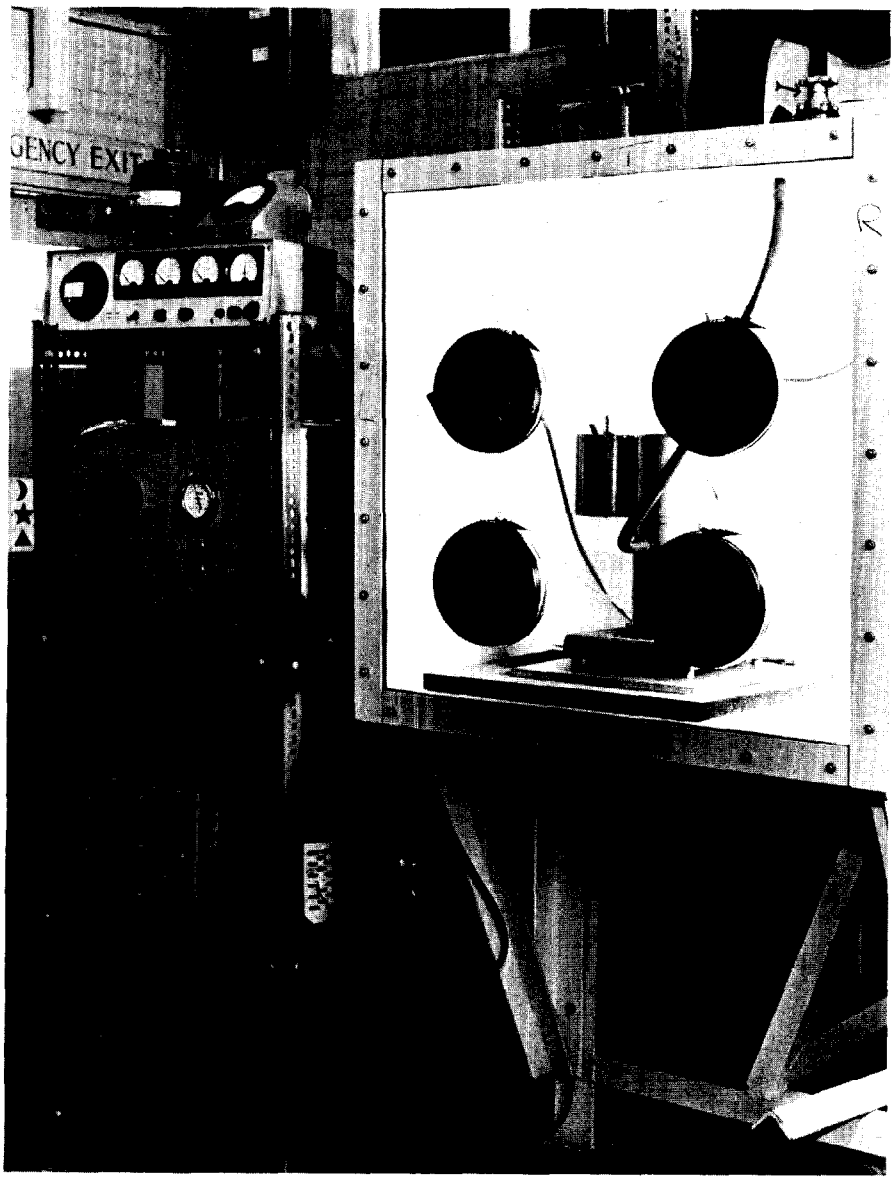

FIGURE 4. GOETZ SPECTROMETER IN GLOVE BOX FOR Pu AEROSOL WORK

Currently a set of experiments, the final in the series, is being carried out aimed at measuring the filterability of plutonium oxide aerosols by medium and high efficiency filter media as a function of gas velocity and particle size. The higher specific activity of plutonium enables the size distributions of these aerosols to be determined with the Goetz spectrometer (Figure 4) using α count techniques.

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DISCUSSION

MURROW: Is there any particular reason why you normally run your air downwards through the filters instead of horizontally? Is it a function of the construction of the filters? Question number two has to do with the cycling equipment, where you circulate the gas through the filter. Is it air or is it inert gas? There has been at least one instance in this country wherein the circulating gas was air; as the temperature went up the decomposition products mixed with air ultimately caused a small explosion as the temperature increased. I was wondering if you used air in your system?

DYMENT: Thank you. The first question on the orientation of the filter pack. This is installed to have the air flow directly downwards in order that particles once deposited on the filter should not drop off during subsequent filter changing operations. The second question on the use of air or inert gases. We used air in this case. There is no, or virtually no, organic material used in the construction of these units so that virtually no decomposition products should be formed. There may in fact be a small amount of organic binder used in the formulation of the paper. The quantity is small and insufficient to cause a build up of combustible gases. We do in fact type test each new brand of filter to check on this very point and if as it sometimes happens we find a lot of smoke produced we know straight away that there are excessive quantities of organics present.

ALVAREZ: I have a question related to the material that you use in your duct work and in the casing of the filter. I noticed that you showed some shots indicating temperature variations around the duct work. Is this painted steel, or is it galvanized?

DYMENT: In general, ducting is either galvanized or painted with a chlorinated rubber paint.

ALVAREZ: What about the joints? I noticed that there were some flanged joints. Is there any particular material for sealing this joint?

DYMENT: Normally, we use neoprene rubber. I wouldn't say that there would be no leakage at 500°C, this is most unlikely. However, we think this is not an important point as the ducting is under suction at all times and should there be some leakage, this would be inward leakage, not outward.

FIRST: I understand from your discussion that the heating tests involved heating the filters and then cooling them before testing them. Is this correct?

DYMENT: This is correct.

FIRST: Have you done any testing of filter efficiency at high temperature on the assumption that in the course of an accident which develops a great deal of heat, you will be very much interested in what these filters will pass during that period.

DYMENT: This is an interesting point. We have run some tests on small samples of certain materials up to about 400°C but we have done no tests on manufactured units. The work which we have done has shown that first of all that the sodium flame test is suitable for use up to about 400°C. And work from other establishments has shown that in fact the efficiency of glass fiber materials to submicron particles does increase rather than decrease, presumably due to improved diffusion factors at the high temperatures.

FIRST: That's not the issue.

DYMENT: I agree; we have not actually carried out any tests on full size equipment at elevated temperatures. All of our efficiency tests on full size equipment are on the basis of carrying out the efficiency tests on the units in the cool state.

CRAIG: I understand from your diagrams that you define breakdown as the point at which penetration increases beyond .05%. If this is the case did you determine why the penetration increased? What was the mode of leakage, if I may put it that way? Was it due to a failure of the filtration material or due to a break away of the filtration material from the metal filter frame or what?

DYMENT: In our opinion the reasons were loss of resilience of the spacers and sealing material, caused by annealing at the high temperature and with repeated heatings and coolings, gaps opened up between the pack and the frame. We don't consider that it was a basic failure of the filter medium itself.

CRAIG: This makes Dr. First's question all the more important, because it seems then that if leakage is due to break away between the medium and the housing this would likely to be more serious at high temperatures than once the filter cooled down again. Or would you not expect this to be the case?

DYMENT: I think it is difficult to say whether in fact a gap once opened up would be further increased when the components contract upon cooling or, it depends upon the way you look at it. I certainly agree that it could have an effect on the efficiency.

OWEN: At these elevated temperatures do you have any chemicals or fumes from these hoods which cause deterioration of these filters which you have tested?

DYMENT: In brief, the answer is no. When we have a problem of fumes and chemicals we endeavor to suppress the source as far as possible in cooperation with the people who are producing the fumes. We ask them to limit their production and try to put some sort of pre-scrubber to prevent the fumes going into the main filter system.

GILBERT: Mr. Dyment, can you tell me what the length of the cycling period was at which you carried up the things in the heated air bath? Did you have these filters in and hooked up with air going through them, or were they merely bathed in the heated air in the cycle?

DYMENT: In the case of what I term the static tests, the filter units were placed in an oven already brought up to temperature. The rate of heating was very rapid and they were subjected to this temperature for a period of an hour and then removed and allowed to cool in the open air. Conditions were slightly different in the case of the recirculating tests in that the oven was not preheated. It took perhaps a quarter of an hour to set the temperature and the filter was then withdrawn from the hot oven and allowed to cool.

GILBERT: Was the air taken through the filters, or were they just immersed in recirculating air?

DYMENT: They were immersed in the air which was pushed through the filters. We had a diaphragm baffle that was constructed and placed inside of the oven in such a way that the recirculating air was forced through the filters.

GILBERT: What was the material of the seal of the filters that you had in here that go up to 500°C? Was this fire clay cement, fiber glass mats, or a combination of both?

DYMENT: When you say seals, do you mean gaskets or the seals around the frame between the pack and the frame?

GILBERT: The frame seal.

DYMENT: These are of a type which are put together without the use of an adhesive such as rubber cement, or fire clay, the seal was made by means of glass fibre.

DOMNING: I notice that you have 20-foot separation between your pre-filter and your HEPA filter. Is that a heat exchanger?

DYMENT: It is acting as a heat exchanger.

DOMNING: Ok. Then if it is, what maximum temperature would you expect to see at your pre-filter should you have a fire in a glove box directly below it or wherever it is located?

DYMENT: We design the system such that there are sufficient heat losses to bring the temperature at the main filter down to 500°C. At the pre-filter, it may of course be higher. In this case, we accept that the pre-filter may be lost.

DOMNING: What would your temperature be at your pre-filter?

DYMENT: Maximum temperature which we allow from, for example, a solvent fire would be about 1000°C. A sufficient length of ducting would be allowed between the pre-filter and the final filter of the system for the temperature to be dropped to 500°C by simple heat exchange.

WILHELM: When you tested your filter element prior to this high temperature treatment by the oil plume test could you see small penetrations like pinholes? Was this test used on those filter elements?

DYMENT: We did not carry out the tests of that type; we made only efficiency tests and pressure drop checks. We did not carry out a pinhole test.

WILHELM: I see. We tried to find filter elements that were tight in the first place before we get high temperatures and we couldn't find one brand which wouldn't show up with pinholes or small leaks, mostly in the cement for sealing between the mat and the frame. We found this only on filter elements for high temperatures. Normal HEPA filter elements for low temperatures wouldn't show pinholes, but on filter elements specially made for high temperatures, 80 - 90% of those filters tested showed pinholes or small leakes in the oil plume test. Relations between the oil plume test and the DOP test for removal efficiency are not finally established, so it is a qualitative test, not a quantitative test. But from experience, we have learned that most of those filter elements, which showed pinholes or small leaks in the oil plume test failed in the installation. We were able to measure activity downstream of those filter elements. I would be interested in learning also if you examinated your filter elements after the heat tests to see what happened to them. Are there holes or cracks and is most of the problem connected to the cement for sealing between mat and frame?

DYMENT: We did not shake the filters after the test. Regarding pinholes, work has been done on the correlation of visible leakages with efficiency measurements, by Fahrbach at the Bonn Staubforschungs Institut, and I believe it is still continuing.